Effect of conductive carbon black on the lightning strikes resistance of carbon fiber-reinforced epoxy resin

Dariusz Krajewski[1),*)] (ORCID ID: 0000-0001-9040-7143), **Mariusz Oleksy**[1)] (0000-0001-5515-8575), **Rafał Oliwa**[1)] (0000-0003-1319-6199), **Grzegorz Masłowski**[2)] (0000-0003-0028-1319), **Kamil Filik**[2)] (0000-0002-6410-3573), **Grzegorz Karnas**[2)] (0000-0002-2255-5926)

DOI: https://doi.org/10.14314/polimery.2024.7.3

Abstract: The effect of conductive carbon black (0.5 wt%) on the properties of carbon fiber-reinforced epoxy resin (Rockwell hardness, Charpy impact strength, tensile and flexural properties, electrical conductivity, and resistance to lightning discharges) was investigated. The composites were obtained by the infusion method. A slight decrease in flexural modulus was observed, while the hardness and Young's modulus increased. The resistivity decreased four times. Simulated multiple lightning discharges confirmed the better electrical conductivity of the composite with the addition of conductive carbon black, which resulted in five times decrease in the laminate damage area.

Keywords: composites, epoxy, carbon black, impulse current, electrical conductivity.

Wpływ sadzy przewodzącej na odporność żywicy epoksydowej wzmocnionej włóknem węglowym na uszkodzenia spowodowane wyładowaniami atmosferycznymi

Streszczenie: Zbadano wpływ sadzy przewodzącej (0,5% mas.) na właściwości żywicy epoksydowej wzmocnionej włóknem węglowym (twardość Rockwella, udarność Charpy'ego, właściwości mechaniczne przy rozciąganiu i zginaniu, przewodnictwo elektryczne i odporność na wyładowania atmosferyczne). Kompozyty otrzymano metodą infuzji. Zaobserwowano nieznaczne zmniejszenie modułu sprężystości przy zginaniu, przy jednoczesnym zwiększeniu twardości i modułu Young'a. Rezystywność zmniejszyła się 4-krotnie. Symulowane wielokrotne wyładowania atmosferyczne potwierdziły lepszą przewodność elektryczną kompozytu z dodatkiem sadzy przewodzącej, co przełożyło się na 5-krotne zmniejszenie obszaru uszkodzenia laminatu.

Słowa kluczowe: kompozyty, żywica epoksydowa, sadza, prąd impulsowy, przewodność elektryczna.

Nowadays, polymer composites are finding more applications as construction materials. Composites are replacing conventional materials such as metal alloys due to their high strength-to-weight ratio, as well as higher corrosion and chemical resistance. However, because of their low electrical conductivity, they cannot be used for effective lightning protection. This feature is particularly important for the aerospace industry due to the high exposure to lightning strikes on aircraft, where materials with low electrical conductivity can be severely damaged [1–4]. There is also a risk of damage to electrical and electronic equipment working inside the flying object due to the presence of the strong electromagnetic field generated by the lightning current flowing through the surface of an aircraft's frame [5].

Composite materials are currently being developed to obtain hybrid composites with improved properties in specific areas such as: increased ballistic resistance [6–8], increased thermal conductivity [9, 10], or increased electrical conductivity [11–40]. Epoxy resin/carbon fiber (EP/CF) composites are most used as a base for improving electrical conductivity due to their high strength and good immunity to lightning discharge, even without modifications [11–14], but this composite has significantly worse electrical properties than metals. Therefore, it is necessary to use various modifications to increase EP/CF electrical conductivity. Literature reviews suggest that the effective modifications can achieved both in the reinforcement and matrix of composites [15–18]. Modifications of the reinforcement are carried out by applying a layer

1) Department of Polymer Composites, Rzeszow University of Technology, Al. Powstańców Warszawy 6, 35-959 Rzeszów, Poland.
2) Department of Electrical and Computer Engineering Fundamentals, Rzeszow University of Technology, ul. W. Pola 2, 35-959 Rzeszów, Poland.
*) Author for correspondence: d.krajewski@prz.edu.pl

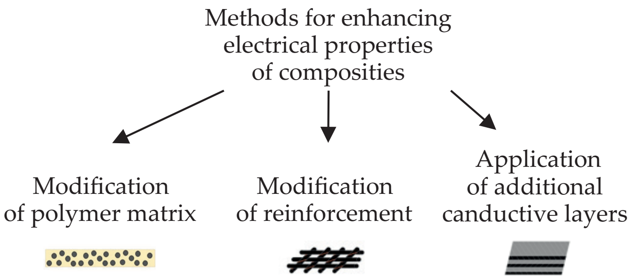

Fig. 1. Methods of modifying electrical properties of polymer composites

of conductive materials [19–27]. To improve the conductivity of the polymer composite matrix, carbon nanofillers such as graphene, graphite, carbon nanotubes or carbon black are most used [28–38]. It is possible to use them as modifiers to the epoxy matrix and as an additional layer with increased conductivity, which is incorporated into the laminate structure (Fig. 1).

Modification of epoxy composites to improve electrical conductivity is an interesting area of research due to the wide range of methods that can be used. Therefore, the aim of this work was to modify EP/CF composite with an optimal filler content selected in previous studies and compare its properties with an unmodified epoxy composite. The results presented in this paper are a continuation of previous studies [39, 40].

EXPERIMENTAL PART

Materials

Epidian 624 epoxy resin (EP), with a density of 1.11 g/cm^3 and an epoxy number of 0.485–0.51 mol/100 g, with Z1 liquid amine hardener supplied by Sarzyna Chemical Ltd. (Nowa Sarzyna, Poland), was used as the matrix of the composites. The matrix modification was carried out with the addition of conductive carbon black (CB) Chezacarb AC supplied by Orlen Unipetrol RPA s.r.o. (Litvinov, Czech Republic). Carbon fiber (CF) of 220 g/cm^3 supplied by Rymatex Ltd. (Rymanow, Poland) was used as reinforcement.

Preparation of composites

Epoxy resin modified with 0.5 wt.% CB (EP_0.5%CB/CF) was obtained by the infusion method. Homogenization of polymer blends was carried out using a high-speed homogenizer Dispermat D-51580 (Reichshof, Germany) with a turbine mixer at 6000 rpm and Hielscher UP400s ultrasounds (Teltow, Germany). Composites reinforced with five layers of carbon fabric were made by the infusion method. The Z1 hardener was used to cross-link the EP matrix in a weight ratio of 100:13. Composites in the form of 300 × 300 × 1.5 mm plates were obtained by the infusion method (Fig. 2) and tested for resistance to lightning current. Samples for mechanical tests were cut from the plates in accordance with the relevant standards. For property comparison, unmodified EP/CF composites were obtained in the same manner.

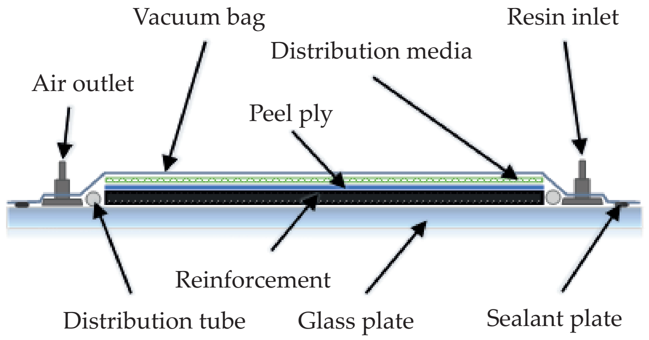

Fig. 2. Scheme of the composite manufacturing system using the infusion method

Methods

Mechanical properties

Rockwell hardness test was carried out using a Zwick/Roell 3106 hardness tester (Ulm, Germany). For each composite, ten measurements were carried out in accordance with ISO 6508. The fracture toughness of the material under dynamic loading was determined using a PSW Gehard Zorn impact test hammer by using the Charpy method, for kinetic energy of impact equal to 1 J. The test specimens were prepared according to EN ISO 179-1, and five measurements were taken for each composite. The static tensile properties were determined using an Instron 5967 (Opole, Poland) testing machine at a croshead speed of 2 mm/min, and 23°C and 50% humidity. The specimens were prepared according to PN-EN ISO 527-4, and five measurements were carried out for each composite. The three-point bending test was carried out on an Instron 5967 (Opole, Poland) three-point contact machine at a crosshead speed of 1 mm/min. The specimens were prepared according to PN-EN ISO 14125, and five measurements were carried out for each composite.

Low-voltage electrical resistivity measurements

Electrical resistivity measurements were performed using the 4-point linear probe method (4P) and the Van der Pauw method (VdP) [40]. The 4P method uses four probes arranged linearly on the sample surface. The outer electrodes are used to force the current flow, and the inner electrodes are used to measure the DC voltage drop. The test uses gold-plated bronze probes in the form of spring needles. The probes arranged symmetrically along one of the symmetry axes, each at 60 mm, were mounted in a plywood cover with dimensions of 300 × 300 mm. The pins touched the surface of the tested samples with a force of 2 N to ensure stable and repeatable measurement results. The measurements were perfor-

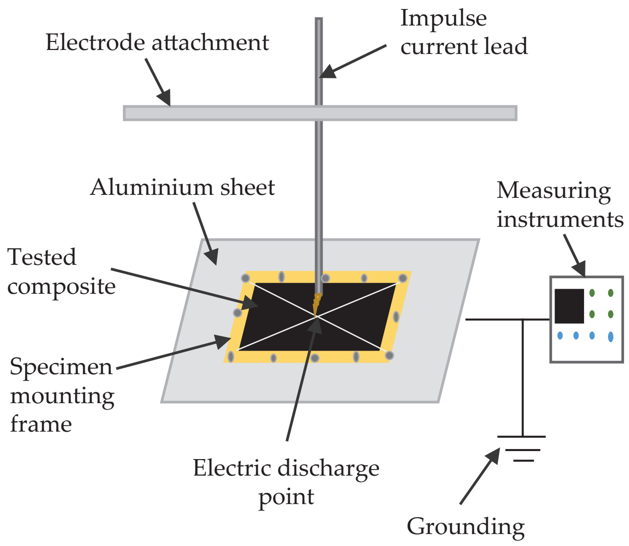

Fig. 3. System for testing the resistance of polymer composites to lightning discharges

med on both surfaces of the samples, which were marked as the upper and lower (bottom - on the glass side of the infusion process).

The Van der Pauw (VdP) method is based on the previously described 4-electrode method. In the 4P technique, the linear electrode arrangement determines the resistivity only in the direction of the probes, while the VdP technique allows determining the average resistivity of the material without knowing the internal structure of the sample. According to the VdP technique, two voltage electrodes and two current electrodes are placed at the edges of the sample. The test setup used the same electrodes as described in the 4P method. Their locations were marked A, B, C, D and correlated with the subsequent arrangement of the panels during the impact tests.

Measurement method with high-current pulses

A test stand was prepared to measure the electrical conductivity, surge resistance and damage resistance of laminates using a pulse generator simulating lightning return-stroke current (Fig. 3).

The generator used can be charged with voltages in the range of 10–80 kV, and the maximum energy stored in the capacitor bank was 10 kJ. After recording the four transient currents $i_A(t)$, $i_B(t)$, $i_C(t)$, $i_D(t)$ flowing from the center of the sample toward each edge, the total current was determined using Equation 1.

$$i(t) = i_A(t) + i_B(t) + i_C(t) + i_D(t) \quad (1)$$

Then, the value of the surge resistance (Rs) was determined as a ratio of the peak value of the impulse voltage *u(t)* to the peak value of the impulse current *i(t)*. An Olympus model DSX510i digital optical microscope (Tokyo, Japan) was used to examine the damage area of the composites after the first current impulse, and then, after four subsequent current impulses (i.e., after five strokes in total).

RESULTS AND DISCUSSION

Mechanical properties

The average values of mechanical properties with standard deviations obtained during testing: Rockwell hardness, Charpy impact strength, tensile and 3-point flexural strength, for the composites: unmodified (EP/CF) and with the addition of 0.5% wt. CB (EP_0.5%CB/CF) are shown in Table 1.

It can be concluded that the addition of CB did not cause significant changes in mechanical properties of EP/CF composite. On the other hand, Rockwell hardness and tensile modulus increased in comparison to EP/CF by 12% and 22%, respectively. Moreover, the deterioration of EP_0.5%CB/CF composite properties in comparison to EP/CF was observed only in the case of flexural modulus (approx. 20%). In the case of the remaining properties, the values of standard deviations do not allow to indicate a material with better properties. On the other hand, higher values of deviations occurring in the case of EP_0.5%CB/CF composite may indicate that its structure is less homogeneous in comparison to EP/CF, which may be caused by the uneven filler distribution in the polymer matrix.

Electrical properties

The results of surface and volume resistivity measurements for EP/CF and EP_0.5%CB/CF composites tested by the 4P are shown in Table 2, while these results for the VdP method are shown in Table 3. The site of the tested composite was considered during the test. The results are presented for specimens before and after using high--current pulses.

Analyzing both methods, EP_0.5%CB/CF composite showed more than three times lower surface resistivity and about four times lower volume resistivity compared to EP/CF composite. For EP_0.5%CB/CF composite, similar

T a b l e 1. Mechanical properties of composites

Composite	Rockwell hardness, N/mm^2	Charpy impact strength, kJ/m^2	Flexural strength MPa	Flexural modulus GPa	Tensile strength MPa	Tensile modulus GPa
EP/CF	110.2±13.1	35.4±1.1	238±43	39.0±2.0	349±14	53±2
EP_0.5%CB/CF	123.8±17.9	34.1±1.7	236±49	31.2±2.4	351±19	65±2

Table 2. Surface and volume resistivity of composites determined by the 4P method before and after high-current impulse testing

Composite	Tested side	Surface resistivity, mΩ		Volume resistivity mΩ·cm	
		Before test	After test	Before test	After test
EP/CF	TOP	526.7	108.6	79.32	16.35
	BOTTOM	1932	175.7	291.1	26.46
EP_0.5%CB/CF	TOP	79.23	81.09	9.666	9.893
	BOTTOM	80.47	77.76	9.818	9.487

T a b l e 3. Surface and volume resistivity of composites determined by the VdP method before and after testing using high-current pulses

Composite	Tested site	Surface resistivity, mΩ		Volume resistivity, mΩ·cm	
		Before test	After test	Before test	After test
EP/CF	TOP	198.50	60.85	29.89	10.43
	BOTTOM	203.10	70.01	30.59	10.54
EP_0.5%CB/CF	TOP	60.84	60.51	7.726	7.684
	BOTTOM	60.85	60.28	7.728	7.656

resistivity was obtained both before and after the impact test, while for the EP/CF composite the differences are significant. About three times lower values were obtained for both quantities. EP/CF composite also showed significant differences before the impact test, when its sites were tested using the 4P method for both surface resistivity (526.7 mΩ top and 1932 mΩ bottom, difference 1405.3 mΩ) and volume resistivity (79.3 mΩ·cm top and 291.1 mΩ·cm bottom, difference 211.8 mΩ·cm). However, after the high-current pulse test, the differences for the EP/CF composite were significantly reduced to 70.1 mΩ for surface resistivity and 10.1 mΩ·cm for volume resistivity, respectively. In contrast, for EP_0.5%CB/CF composite, there were no significant differences in properties due to the laminate site that was tested both before and after the test.

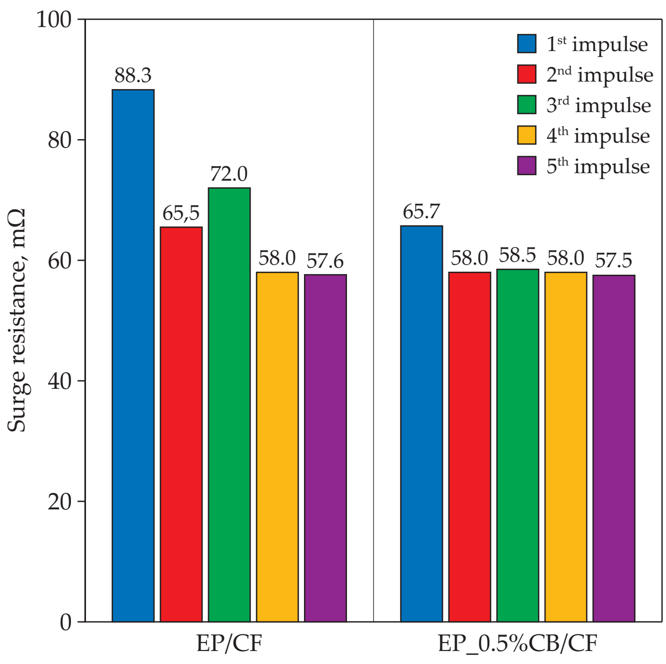

Fig. 4. Surge resistance of composites for successive current pulses

Figure 4 shows the impedance measured during the high-current test. The measurements were taken during an electrical impulse using an oscilloscope and then converted to the surge.

The surge resistance was calculated for all samples based on the voltage and current waveforms measured during the test with each of five consecutive current pulses. It can be seen a decrease in the obtained values for both composites at the first stroke, while with the action of the subsequent strokes the analyzed surge resistances of the two composites becomes more and more equal. Due to the burning of the matrix in the action of the first current pulse, an area with a well-conductive carbon fiber was exposed, which can be observed in the images shown below. Through this, the subsequent pulses to which the samples were subjected hit an area with similar properties, due to the use of the same material as reinforcement for both composites tested. The most significant difference, however, can be seen in the results obtained for the first current pulse, where for the EP_0.5%CB/CF composite the value of the surge resistance was lower by about 25% compared to EP/CF. This property allows for a more effective dissipation of the electrical charge at the very first contact of the electrical discharge with the surface of the laminate, which reduces the area of its damage and the risk of induced dangerous overvoltage's in electrical and signal cable bundles.

Damage surface analysis

The images of EP/CF and EP_0.5%CB/CF composites after applying five pulses simulating a multi-strike lightning flash are shown in Fig. 5.

The estimated damage area of the laminate surface was about five times smaller for EP_0.5%CB/CF composite than for EP/CF composite. This confirms the previously presented conclusions regarding the impact resistance for the first current pulse. The example of the

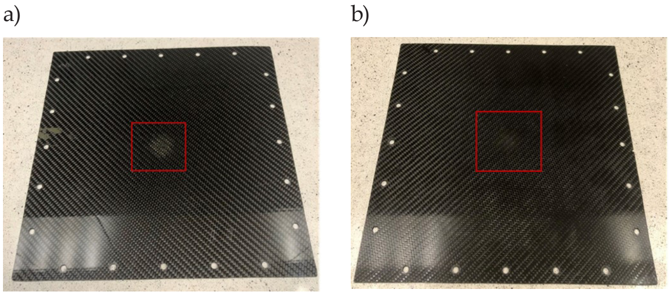

Fig. 5. Photographs of the surface after impact testing of composites: a) EP/CF, b) EP_0.5%CB/CF

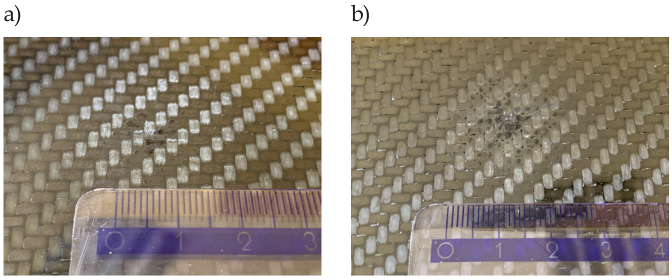

Fig. 6. Photographs of damage areas of EP_0.5%CB/CF composite: a) after one impulse, b) five impulses

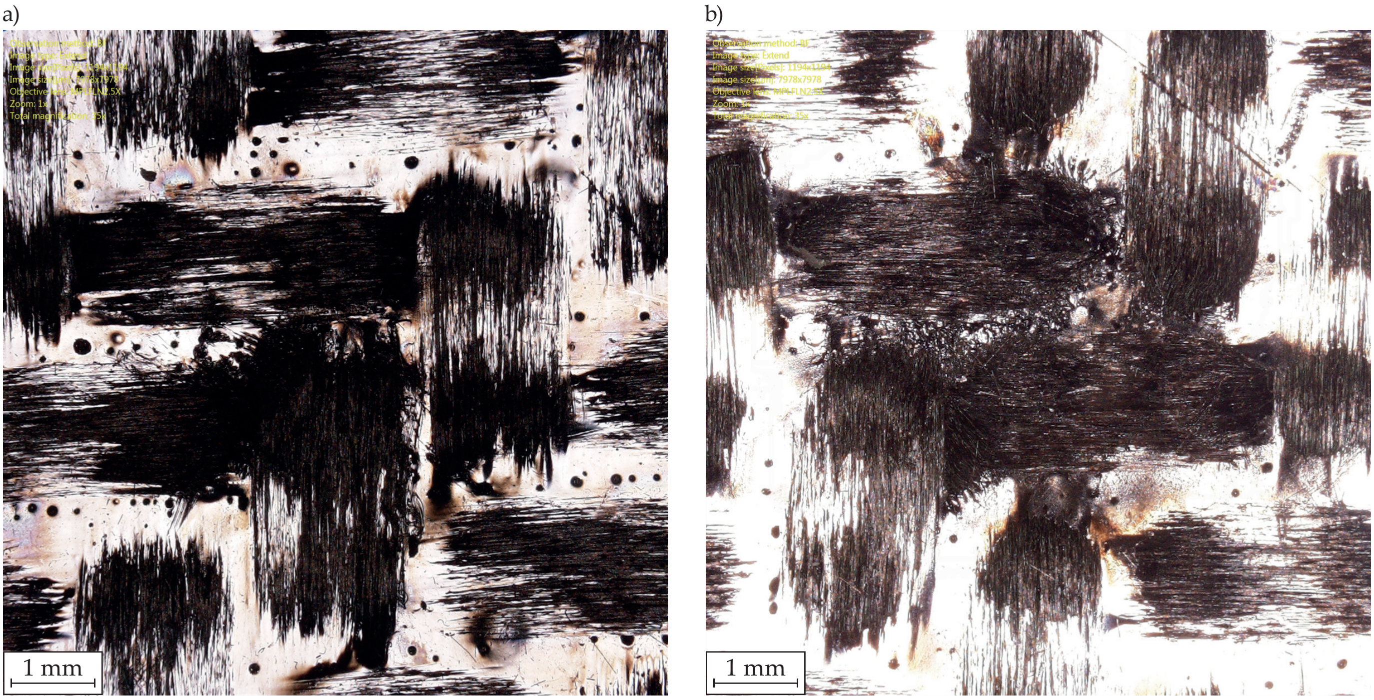

Fig. 7. Microscopic images of the damage center of EP_0.5%/CB/CF composite: a) one impulse, b) five impulses; 35× magnification

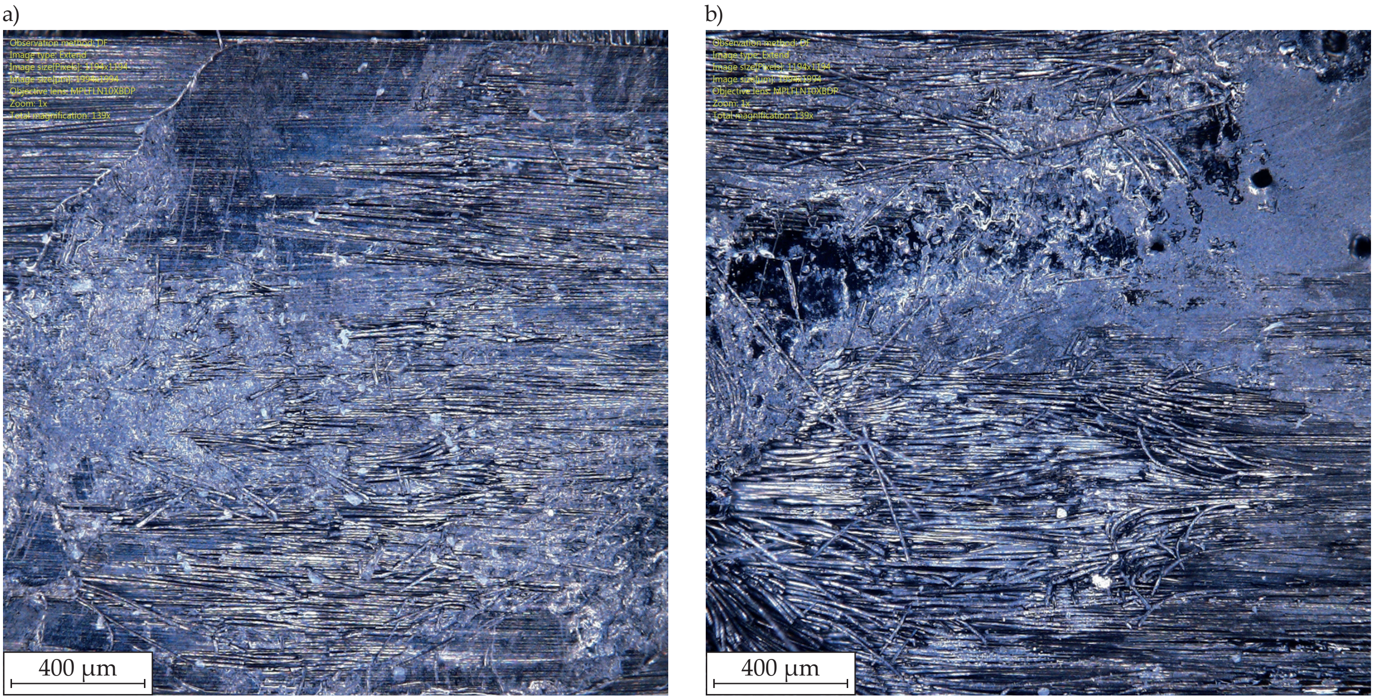

Fig. 8. Microscopic images of the damage center of EP_0.5%/CB/CF composite: a) one impulse, b) five impulses; 139× magnification

EP_0.5%CB/CF composite shows the differences in the structure of damage occurring at the site of the electric discharge after the first and fifth pulse (Fig. 6).

A significant increase in the damage area can be seen for the composite after the fifth pulse, compared to the first pulse it has about three times more the area. However, fully exposed fragments of CF reinforcement are visible only in the central area of the electric discharge strike. The farther away from the epicenter of the impact, the exposure of the fibers compared to the undamaged matrix is much smaller.

Fig. 7 anf 8 show the centers of the electrical discharge areas after 1 and after 5 pulses for EP_0.5%CB/CF composite observed under an optical microscope at 35× and 139× magnification.

Based on the microscopic images, it is possible to observe an increase in the ratio of exposed carbon fibers also in the center of the damage, for the composite subjected to five current pulses. The 139× magnification images (Fig. 8) also allows to observe that a part of the fiber surface band, located in the very center of the impact after the fifth pulse, was interrupted. This indicates a large current flow in this area, which can melt some of the carbon fiber bands, which can weaken the mechanical properties of the composite at the lightning discharge site.

CONCLUSIONS

Carbon fiber reinforced epoxy composites were obtained by infusion method. Conductive carbon black (0.5 wt%) was used to improve electrical properties of the polymer matrix. The addition of carbon black improved hardness and tensile modulus and decreased flexural modulus, without a significant effect on other mechanical properties. Low voltage 4P and VdP conductivity tests showed that the resistivity of the carbon black modified composite was on average four times lower compared to EP/CF composite. On the other hand, tests with current pulses simulating a multi-surge lightning flash confirmed the achievement of better electrical conductivity for the EP_0.5%CB/CF composite even under the action of the first electrical discharge pulse, which reduces the risk of induced dangerous overvoltage's in electrical and signal cable bundles. Tests of the laminate surface at the discharge point showed a 5-fold reduction in the damage area for the carbon black modified composite. Such a significant reduction reduces the probability of complete burnout of the composite at the point of the electrical impulse. This parameter is particularly important, among others, in the case of the use of materials as structural elements of aircraft, which are exposed to high stresses during operation.

Authors contribution

D.K. – writing-original draft, conceptualization, investigation, visualization; M.O. – supervision, writing-review and editing; R.O. – methodology; G.M. – formal analysis, writing-review and editing; K.F. – resources; G.K. – validation. All authors have read and agreed to the published version of the manuscript.

Funding

This research received no external funding.

Conflict of interest

The authors declare no conflict of interest.

REFERENCES

[1] Gagne M., Therriault D.: *Progress in Aeronautical Sciences* **2014**, *64*, 1. https://doi.org/10.1016/j.paerosci.2013.07.002

[2] Willett J.C., Park G., Krider E.P. *et al.*: "Triggered lightning risk assessment for reusable launch vehicles at the southwest regional and Oklahoma spaceports" Materials from 86th AMS Annual Meeting, Atlanta, Georgia, January 2006.

[3] Pavan C., Fontanes P., Urbani M. *et al.*: *Journal of Geophysical Research: Atmospheres* **2020**, *125*, 1. https://doi.org/10.1029/2019JD031245

[4] Malinga G.A., Niedzwecki J.M.: *Renewable Energy* **2016**, *87*, 572. https://doi.org/10.1016/j.renene.2015.10.047

[5] Paolone M., Rachidi F., Borghetti A. *et al.*: *IEEE Transactions on Electromagnetic Compatibility* **2009**, *51*, 532. https://doi.org/10.1109/TEMC.2009.2025958

[6] Hua F., Su1 L., Luo X. *et al.*: *Journal of Physics: Conference Series* **2023**, *2478*, 072072. https://doi.org/10.1088/1742-6596/2478/7/072072

[7] Czech K., Oleksy M., Oliwa R. *et al.*: *Polimery* **2022**, *67*, 552. https://doi.org/10.14314/polimery.2022.11.2

[8] Zhang R., Han B., Zhong J.Y. *et al.*: *International Journal of Impact Engineering* **2022**, *159*, 104035. https://doi.org/10.1016/j.ijimpeng.2021.104035

[9] Liu H., Chen H., Chen H. *et al.*: *Journal of Applied Polymer Science* **2024**, *e55866*. https://doi.org/10.1002/app.55866

[10] Sun X., Pan X., Wu Y. *et al.*: *Industrial & Engineering Chemistry Research* **2024** *63*, 11030. https://doi.org/10.1021/acs.iecr.4c012

[11] Wang Y., Jing X.: *Polymers for Advanced Technologies* **2005**, *16*, 344.

https://doi.org/10.1002/pat.589
[12] Katunin A., Krukiewicz K., Herega A. *et al.*: *Advances in Materials Science* **2016**, *16*, 32. https://doi.org/10.1515/adms-2016-0007
[13] Feraboli P., Miller M.: *Composites, Part A: Applied Science and Manufacturing* **2009**, *40*, 954. https://doi.org/10.1016/j.compositesa.2009.04.025
[14] Soutis C.: *Materials Science and Engineering: A* **2005**, *412*, 171. https://doi.org/10.1016/j.msea.2005.08.064
[15] Krajewski D., Oleksy M., Oliwa R. *et al.*: *Energies* **2022**, *15*, 4562. https://doi.org/10.3390/en15134562.
[16] Atif R., Shyha I., Inam F.: *Polymers* **2016**, *8*, 281. https://doi.org/10.3390/polym8080281
[17] Ogbonna V.E., Popoola A.P.I., Popoola O.M. *et al.*: *Polymer Bulletin* **2022**, *79*, 6857. https://doi.org/10.1007/s00289-021-03846-z
[18] Kashfipour M.A., Mehra N., Zhu J.: *Advanced Composites and Hybrid Materials.* **2018**, *1*, 415. https://doi.org/10.1007/s42114-018-0022-9
[19] Bao D., Gao Y., Cui Y. *et al.*: *Chemical Engineering Journal* **2022**, *433*, 133519. https://doi.org/10.1016/j.cej.2021.133519
[20] Kim H.S., Hahn H.T.: *AIAA Journal* **2009**, *47*, 2779. https://doi.org/10.2514/1.39522
[21] Wu X., Tang B., Chen J. *et al.*: *Composites Science and Technology* **2021**, *203*, 108610. https://doi.org/10.1016/j.compscitech.2020.108610
[22] Phua J.L., Teh P.L., Ghani S.A. *et al.*: *Polymers for Advanced Technologies* **2017**, *28*, 345. https://doi.org/10.1002/pat.3894
[23] Radouane N., Depriester M., Maaroufi A. *et al.*: *Journal of the Chinese Chemical Society* **2021**, *68*, 1456. https://doi.org/10.1002/jccs.202000490
[24] Zhang D., Ye L., Deng S. *et al.*: *Composites Science and Technology* **2012**, *73*, 412. https://doi.org/10.1016/j.compscitech.2011.12.002
[25] Gungor S., Bakis C.E.: *Journal of Composite Materials* **2015**, *49*, 535. https://doi.org/10.1177/0021998314521256
[26] El-Tantawy F., Kamada K., Ohnabe H.: *Journal of Applied Polymer Science* **2003**, *87*, 97. https://doi.org/10.1002/app.10851
[27] Macutkevic J., Kuzhir P., Paddubskaya A. *et al.*: *Journal of Applied Physics* **2013**, *114*, 033707. https://doi.org/10.1063/1.4815870
[28] Zhang H., Liu Y., Kuwata M. *et al.*: *Composites Part A: Applied Science and Manufacturing* **2015**, *70*, 102. https://doi.org/10.1016/j.compositesa.2014.11.029
[29] Rehbein J., Wierach P., Gries T. *et al.*: Composites Part A: Applied Science and Manufacturing **2017**, *100*, 352. https://doi.org/10.1016/j.compositesa.2017.05.014
[30] Han J.H., Zhang H., Chen M.J. *et al.*: *Carbon* **2015**, *94*, 101. https://doi.org/10.1016/j.carbon.2015.06.026
[31] Kawakami H., Feraboli P.: *Composites Part A: Applied Science and Manufacturing* **2011**, *42*, 1247. https://doi.org/10.1016/j.compositesa.2011.05.007
[32] Zhang B., Soltani S.A., Le L. *et al.*: *Materials Science and Engineering: B* **2017**, *216*, 31. https://doi.org/10.1016/j.mseb.2017.02.008
[33] Kumar V., Sharma S., Pathak A. *et al.*: *Composite Structures* **2019**, *210*, 581. https://doi.org/10.1016/j.compstruct.2018.11.088
[34] Munalli D., Dimitrakis G., Chronopoulos D. *et al.*: *Composites Part B: Engineering* **2019**, *173*, 106906. https://doi.org/10.1016/j.compositesb.2019.106906
[35] Misiura A.I., Mamunya Ye.P., Kulish M.P.: *Journal of Macromolecular Science, Part B: Physics* **2020**, *59*, 121. https://doi.org/10.1080/00222348.2019.1695820
[36] Zhang J., Zhang X., Cheng X. *et al.*: *Composites, Part B: Engineering* **2019**, *168*, 342. https://doi.org/10.1016/j.compositesb.2019.03.054
[37] Wang B., Duan Y., Xin Z. *et al.*: *Composites Science and Technology* **2018**, *158*, 51. https://doi.org/10.1016/j.compscitech.2018.01.047
[38] Zhao Z., Ma Y., Yang Z. *et al.*: *Materials Today Communications* **2020**, *25*, 101502. https://doi.org/10.1016/j.mtcomm.2020.101502
[39] Filik K., Karnas G., Masłowski G. *et al.*: *Energies* **2021**, *14*, 7899. https://doi.org/10.3390/en14237899
[40] Filik K., Oliwa R., Karnas G. *et al.*: *Measurement* **2022**, *199*, 111546. https://doi.org/10.1016/j.measurement.2022.111546

Received 28 V 2024.
Accepted 21 VI 2024.