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CRYOGENIC TEMPERATURE INFLUENCE ON COMPOSITE MATERIAL BEHAVIOUR

UTICAJ KRIOGENE TEMPERATURE NA PONAŠANJE KOMPOZITNOG MATERIJALA

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- 3D printing
- cryogenic environment
- structural integrity

Ključne reči

- kompoziti
- 3D štampa
- kriogeni uslovi
- integritet konstrukcije

Abstract

The purpose of this study is to compile the published literature dealing with the impact of cryogenic temperature on the behaviour of composite materials and suggest 3D printing composites for the same purpose. The behaviour of fibre-reinforced polymer composites under extreme temperature conditions is investigated and effects of temperature on strength, stiffness, ductility, and fracture toughness of the matrix material is explored. Experimental results show that low temperatures can lead to increased strength and Young's modulus, but reduced ductility. The limited existing literature on cryogenic behaviour of 3D printed composite materials highlights the need for new research studies to provide insights into their performance at cryogenic temperatures, particularly for applications in the aerospace industry.

Izvod

Osnovna svrha ovog rada je sagledavanje literature objavljene na temu uticaja kriogenih temperatura na ponašanje kompozitnih materijala i predlog da se za iste uslove može koristiti 3D štampani kompozit. Ovaj rad sagledava ponašanje vlaknasto-ojačanih kompozita pri dejstvu ekstremnih temperaturnih uslova i kakve efekte temperatura ima na čvrstoću, krutost, duktilnost i žilavost loma materijala matrice. Eksperimentalni rezultati su pokazali da niske temperature uzrokuju porast čvrstoće i Jangovog modula, dok je duktilnost redukovana. Ograničeni podaci iz literature na temu uticaja 3D štampanih kompozita naglašavaju potrebu za novim studijama koje bi imale za cilj da pruže detaljniji uvid u njihove performanse u kriogenom okruženju, naročito u primeni svemirskih tehnologija.

INTRODUCTION

Fibre reinforced polymer composites are widely used in many industries due to their excellent properties, such as high strength-to-weight ratio and corrosion resistance. However, there are certain extreme environments both within and beyond the Earth's atmosphere where these composites must perform at cold (cryogenic) temperatures and the usage is mostly in spacecraft and launch vehicle industries, aircraft structures, and Arctic exploration systems. In these extreme environments, the behaviour of composite materials can change significantly, and it is essential to understand the effect of low and cryogenic temperatures on their properties.

A decrease in temperature, either during the manufacturing process or during low-temperature operating conditions, can cause the matrix to shrink. The coefficient of thermal expansion of the matrix is typically an order of magnitude greater than that of the fibres. Consequently, the contraction of the matrix is resisted by relatively stiff fibres through fibre-matrix interface bonding, leading to the establishment of residual stresses within the material microstructure. However, the residual stresses may become sufficiently large to cause micro-cracking within the matrix and at matrix-fibre interfaces. Formation of such micro-cracks can compromise the overall strength and integrity of the composite material. In particular, the development of macroscopic matrix cracks can occur when the density and size of microscopic cracks reach a certain threshold. Changes in temperature can have a profound impact on the strength and stiffness of the matrix material. As most resin materials become stronger and stiffer at lower temperatures, the overall performance of the composite material can be significantly altered.

The cracks in materials are a significant concern for structural integrity because cracks can reduce the load-carrying capacity of a structure. Cracks act as local stress concentrators, which can result in localized yielding and plastic deformation following crack propagation. In the case of brittle materials, such as a polymer matrix in a cryogenic environment, cracks are particularly dangerous. In extreme cases, the structure may collapse due to the failure of the material. Future investigations should focus on filling the knowledge gaps in this area to facilitate practical advancements in aerospace engineering.

LITERATURE REVIEW

To gain a more comprehensive understanding of this particular engineering field, it would be beneficial to provide a summary of some research papers that investigate the properties of fibre-reinforced polymer composites under cryogenic conditions. Such research papers include different testing methodologies, including tensile, bending, impact, fatigue, and thermal cycling tests.

The research conducted by Hunt et al. /1/ investigate the mechanical properties of 3D printed composites at cryogenic temperatures, specifically the ultimate tensile strength of carbon-fibre reinforced PETG (polyethylene terephthalate glycol) and carbon-fibre reinforced Amphora AM1800 filament. In this paper the ultimate tensile strength of carbon-fibre reinforced PETG has increased by 49 % and for carbon-fibre reinforced Amphora AM1800 filament the ultimate tensile strength decreased by 29.9 % at cryogenic temperatures compared to room temperature. The modulus of elasticity of carbon-fibre reinforced PETG increased by 43.2 % at cryogenic temperatures compared to room temperature, from 55.5 to 82.8 MPa, /1/.

The percent elongation at fracture of carbon-fibre reinforced PETG decreased from 2.5 to 1.84 % at cryogenic temperatures compared to room temperature. It can be concluded that PETG is superior to Amphora AM1800 filament in use at cryogenic temperature. The study does not investigate the effect of different printing parameters, or post-processing treatments on the mechanical properties of materials. Also, the study /1/ does not investigate the long-term durability or fatigue behaviour of materials at cryogenic temperatures which can be gap in further research.

Another paper conducted by Diansen et al. /2/ investigates three-point bending tests on 3D integrated woven spacer composites with different core heights at room- and liquid nitrogen temperature, and after that the macro-fracture morphology and SEM (scanning electron microscope) micrographs are used to understand the deformation and failure mechanism of the composites. It was shown that the bending properties at liquid nitrogen temperature are significantly improved compared to those at room temperature, and the bending properties at both room and liquid nitrogen temperature can be greatly affected by the core height. The conclusion in /2/ was that at liquid nitrogen temperature, the material shows brittle fracture characteristics, but the core height is still the main parameter that dominates the bending properties - as height increases, the bending properties also increase. Load-deflection curve at liquid nitrogen temperature declines significantly showing obvious brittle fracture feature and zigzag fluctuations. At room temperature there are different dominant failure mechanisms for each specimen with different core height, but at liquid nitrogen temperature, all composite specimens show identical gradual failure process which extends from the external layers of the compression face to the interior ‘8’ shaped core layers.

Gómez-del Río et al. /3/ examine the response of carbon fibre-reinforced epoxy matrix (CFRP) laminates at low impact velocity and at low temperature conditions. The study used square specimens of carbon fibre/epoxy laminates with different stacking sequences (unidirectional, cross-ply, quasi-isotropic and woven laminates) and were tested using a Drop Weight Tower device. The test temperature ranged from 20 down to -150 °C. After the impact tests, the damage extension was measured by C-Scan ultrasonic inspection and the damage mechanisms were studied by optical and scanning electron microscopy. During microscopic inspection, different types of damages were observed in the CFRP specimens, including matrix cracking, fibre breakage, delamination, and fibre pull-out. The type of damage varied depending on the ply reinforcement architecture and stacking sequence of the laminates. It is observed that the temperature directly corresponds to the diameter and shape of damaged area impacted by striker - as temperature decreases, the diameter of the damaged area increases, /3/.

Kaushik et al. /4/ conducted two different testing regimes, Mode-I and Mode-II interlaminar fracture tests, under extremely cold environmental conditions where specimens were ‘soaked’ in a cold environmental chamber at a test temperature for 40 min before fracture testing to ensure continual temperature field. Mode-I fracture toughness refers to the ability of a material to resist crack propagation under tensile loading, where the crack is perpendicular to the direction of the applied load, and Mode II loading involves applying a force perpendicular to the direction of fibres in the composite material, causing the layers of the material to slide against each other. This type of loading is important to study because it can occur in real-world situations, such as when an aircraft experiences turbulence. It can be concluded that a significant reduction in Mode-I and Mode-II fracture toughness is observed due to the nature of the matrix under cold temperature conditions, /4/.

Another interesting paper by Meng et al. /5/ investigates the effects of cryogenic temperature and thermal cycling (50, 100 and 150 cycles) on the mechanical properties and failure modes of carbon fibre reinforced polymer (CFRP) laminates with different stacking sequences. Static tensile and three-point bending tests were conducted. The sizes of samples were set according to the recommendation of ASTM D3039 and D7264. It is interesting to mention that the influence of thermal cycling on the mechanical properties was found to be insignificant for tensile tests (1.4-2.4 %), and 10 % for bending tests under slow cooling rates of 7 K/min, /5/.

Li et al. /6/ discuss the properties of a special type of plastic called polyethersulfone (PES). The study also examines the effect of adding graphene oxide (GO) and short carbon fibres (SCFs) to PES in order to enhance its cryogenic mechanical properties. The cryogenic temperature for mechanical testing was achieved by dipping samples in liquid nitrogen for over 10 minutes, and at least five specimens were measured for each specimen composition. The results show that the addition of graphene oxide (GO) coating on short carbon fibres (SCFs) is an effective way to improve cryogenic mechanical properties of polyethersulfone (PES) composites, /6/.

Li et al. /7/ report on the development of carbon fibre/epoxy composites with improved mechanical properties by using a flexible polymer (polysiloxane) containing -Si-O-Si- molecular chains and epoxy groups (EPSE). The results showed that the addition of the polysiloxane improved the flexural strength, fracture toughness, and impact strength of the composites at room temperature. The composites also exhibited good cryogenic mechanical properties at 77 K, where tensile strength of EPSE reached 207 MPa which is 17.6 % higher than that of the neat epoxy, and failure strain reached 3.13 % which is 29.3 % higher than neat epoxy, indicating the better flexibility of EPSE-epoxy at cryogenic conditions. At cryogenic temperature epoxy resin has a failure strain much lower than at room temperature, but in case of composite with EPSE content, tensile strength and elastic modulus are much higher than at room temperature. The study suggests that incorporating flexible molecular structures into epoxy curing networks is an effective method for improving the mechanical properties of epoxy resins, /7/.

It is worth mentioning a few additional papers /8-10/ which investigate carbon composite at low temperature, but in the range from -50 to -100 °C.

The research gap can be an investigation into the potential of 3D printing technologies, specifically additive manufacturing, that employ short-reinforced fibres in the production of complex parts that can be used in space engineering. A multitude of production parameters significantly impact the final product, indicating that a great opportunity exists for future researchers to further explore this area. A new study should aim to fill the research gap in this area and provide insights into the behaviour of 3D printed materials at low temperatures which can have practical implications for the aerospace industry.

TESTING OF 3D PRINTED SPECIMEN

In this section, attention is focused on a basic proposition for different testing types of the 3D printed short carbon fibre reinforced composite. There are three relatively simple research experiments which can be done on standard (widely used) equipment: tensile, bending, and impact testing. Charpy impact testing is probably the easiest experiment from the standpoint of available specialized equipment. By the nature of impact testing, the experiment can be completed in short amount of time, so the temperature field of the specimen should not be greatly affected by the temperature change. In /11/ it is stated that after 40-50 min, the impact test was conducted within 5 s for each specimen, due to the time the specimen was brought out from insulated chamber. Also, each case was repeated three times to ensure the reliability of the testing results.

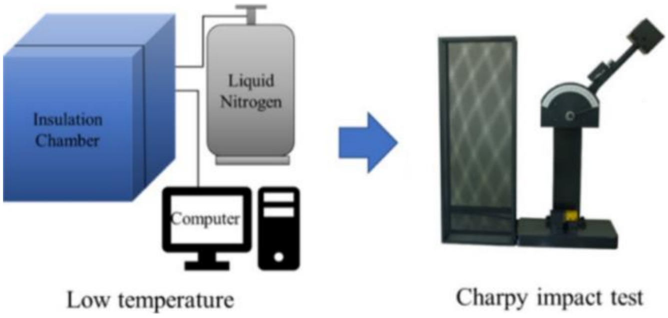


Figure 1. Experimental procedure for impact testing specimens at cryogenic temperature, /11/.

In case of testing at -196 °C the specimen can be directly immersed in liquid nitrogen at atmospheric pressure without the need for an insulated chamber, similarly to reference /12/. In aerospace engineering, structures can be damaged by meteor impacts. When a meteor collides with a spacecraft or some other structure, it can cause impact damage and abrasion. To mitigate the risk, the spacecraft and other structures are typically equipped with protective measures such as shielding, designed to absorb or deflect meteor impacts. It can be concluded that impact testing of complex geometry parts in space engineering (potentially 3D printed) is a real opportunity, and new research should further investigate this field. Bending and tensile tests are the two crucial mechanical tests that provide essential information on the mechanical properties of materials, including those used in cryogenic environments. These tests determine the strength, ductility, and stiffness of materials, and are essential for the design and manufacture of various structural components and products in cryogenic applications. In cryogenic environments, material properties can change significantly due to low temperatures, leading to increased brittleness and reduced ductility. Because testing is not fast enough to avoid great change in temperature (like impact testing), in case of tensile and bending testing it is important to completely immerse specimens in the liquid nitrogen until the test is complete.

Schematic diagrams of the cryogenic testing system for tensile and three-point bend tests can be seen in Fig. 2, where specimens are ‘soaked’ in liquid nitrogen /6/, and in Fig. 3 it can be seen that the bending stiffness increases at cryogenic temperature.

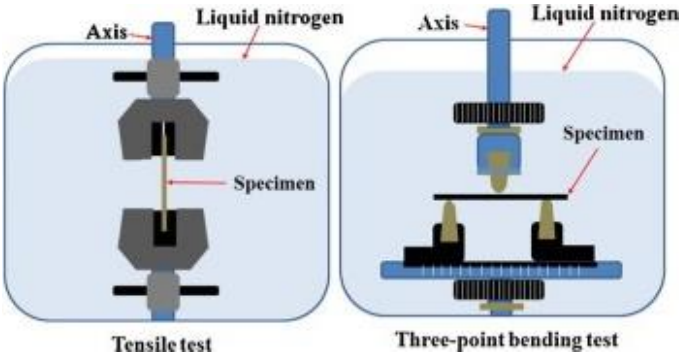
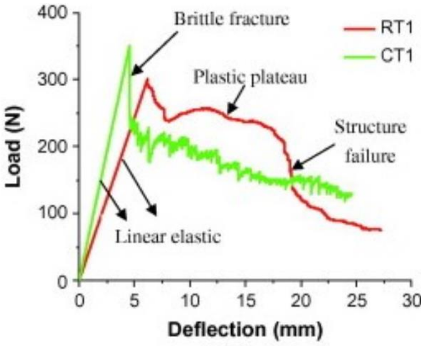


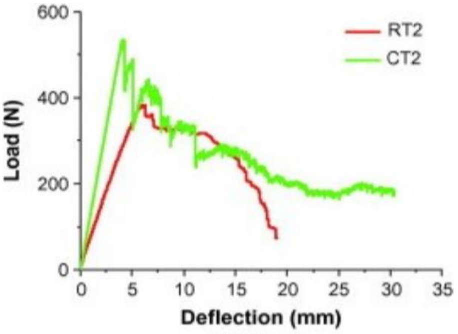
Figure 2. Cryogenic test system for tensile and three-point bend tests where specimens are ‘soaked’ in liquid nitrogen, /6/.

Similarly to research paper /2/, it is possible to make 3D printed composite lattice plate structure to examine three-point bend test and investigate the parameters of the printing angle on the deflection of the plate.

It is crucial to perform both bending and tensile tests in cryogenic environments to accurately evaluate the mechanical properties of materials in these conditions. In bending tests, a 3D printed specimen of the material can be placed on two supports and subjected to a load at the midpoint while being maintained at cryogenic temperatures. The deflection of the material can be measured as the load increases, and maximum stress at failure can be calculated afterwards. Similarly, in case of the 3D printed specimen and tensile testing, the material is pulled in the opposite direction until it fractures, while at cryogenic temperatures. The load and elongation are measured throughout the test, and the stress-strain curve can be plotted. Elongation properties, tensile and yield strength can be obtained from this curve. Bending and tensile tests enable a comprehensive evaluation of the mechanical properties of materials in cryogenic environments and provide useful information for the design and optimisation of structural components subjected to these extreme conditions.



(a) 3mm



(b) 6mm

Figure 3. Bending stiffness increase on cryogenic temperature in case of different core heights: a) 3 mm; b) 6 mm, /2/.

ADVANTAGES AND DISADVANTAGES

One of the primary advantages of 3D printing technology is the ability to produce complex geometries cost-effectively, which is highly desirable in the aerospace industry. Traditional manufacturing processes are often unable to produce parts with complex geometries, requiring multiple manufacturing steps and increasing the cost of production. 3D printing technology can produce complex geometries in a single step, significantly reducing the time and cost of production. This is especially beneficial for the production of small parts that are difficult to produce using traditional manufacturing techniques. Another significant advantage of 3D printing technology in cryogenic environments is the ability to produce components on-demand. Some research papers have shown that properties of composite materials can be improved at cryogenic temperatures.

This approach reduces the need for stockpiling and storage of parts, which can be costly, especially in the space industry. With 3D printing technology, parts can be produced as needed, reducing the amount of space required for storage and the overall cost of production.

However, there are several disadvantages associated with the use of 3D printing technology in cryogenic environments that must be considered. One disadvantage is that not all filaments from different manufacturers have identical properties. This variation in material properties can affect the testing accuracy, making it challenging to conduct standardised testing across various manufacturers. As a result, it is necessary to thoroughly investigate the material properties of each filament and ensure that proper testing procedures are in place to obtain accurate results. Another disadvantage of using 3D printing technology in cryogenic environments is that specialised testing chambers are required for testing specimens immersed in cryogenic fluids. The testing chamber must be designed to withstand extremely low temperatures of the cryogenic environment. Furthermore, 3D printed specimens are highly sensitive to printing parameters, such as printing angle, layer height, and temperature, which can significantly impact the results of the testing procedures. The sensitivity of 3D printed specimens to printing parameters requires extensive testing to derive accurate conclusions. Thorough investigations are necessary to understand the effect of each parameter on the final product and ensure that accurate testing results are obtained.

Cryogenic temperatures can affect sensors used for testing, leading to inaccurate results. The low temperatures can cause sensors to freeze or malfunction, which can significantly impact the accuracy of test results. It is necessary to use sensors specifically designed for use in cryogenic environments to ensure accurate and reliable test results. Optical specimen testing methods such as digital image correlation (DIC) are not suitable for use at cryogenic temperatures. This is because the visibility of the specimen is not optimal in the cryogenic environment, making it challenging to obtain accurate measurements using optical testing methods. This limitation can make it difficult to accurately measure the properties of 3D printed parts in cryogenic environments, making it necessary to rely on other testing methods.

It can be concluded that 3D printing technology has both advantages and disadvantages when used in cryogenic environments. While it can significantly reduce costs and increase efficiency, the sensitivity of 3D printed specimens to printing parameters and the need for specialised testing chambers require thorough investigation and careful consideration of the technology's suitability for specific applications. By understanding the advantages and disadvantages of 3D printing technology, researchers and engineers can make informed decisions about its use in cryogenic environments.

CONCLUSIONS

Fibre-reinforced polymer composites have excellent properties, but they behave differently in extreme environments. Changes in temperature can also impact the strength and stiffness of the matrix material.

It has been observed that these temperatures have effect on composites, leading to enhanced strength and Young's modulus. However, the opposite trend is observed in terms of ductility, with lower failure strain and fracture toughness. Also, thermal cycling on the mechanical properties was found to be insignificant for tensile tests and incorporating flexible molecular structures into epoxy curing can be an effective method for improving the mechanical properties of epoxy resins.

To the authors' best knowledge, there is insufficient literature on the cryogenic behaviour of 3D printed composite materials, so new research studies should aim to provide insights into the behaviour of 3D printed materials at extremely low temperatures which can have practical implications for aerospace industry. To design space structures that can handle these conditions, researchers need to study the material properties, failure mechanisms, and experimental procedures.

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REFERENCES

1. Hunt, M., Salmon, K., Haney, J., et al. (2020), *Ultimate tensile strengths of 3D printed carbon-fibre reinforced thermoplastics in liquid nitrogen*, IOP Conf. Ser.: Mater. Sci. Eng. 755: 012118. doi: 10.1088/1757-899X/755/1/012118
2. Li, D.-S., Zhao, C.-Q., Jiang, L., Jiang, N. (2014), *Experimental study on the bending properties and failure mechanism of 3D integrated woven spacer composites at room and cryogenic temperature*, Compos. Struct. 111: 56-65. doi: 10.1016/j.compstruct.2013.12.026
3. Gómez-del Río, T., Zaera, R., Barbero, E., Navarro, C. (2005), *Damage in CFRPs due to low velocity impact at low temperature*, Compos. Part B: Eng. 36(1): 41-50. doi: 10.1016/j.compositesb.2004.04.003
4. Kaushik, V., Bar, H.N., Ghosh, A. (2021), *Influence of extremely cold environmental conditions on interfacial fracture phenomenon of aerospace grade unidirectional composites*, Thin-Walled Struct. 161: 107431. doi: 10.1016/j.tws.2020.107431
5. Meng, J., Wang, Y., Yang, H., et al. (2020), *Mechanical properties and internal microdefects evolution of carbon fiber reinforced polymer composites: Cryogenic temperature and thermocycling effects*, Compos. Sci. Technol. 191: 108083. doi: 10.1016/j.compscitech.2020.108083
6. Li, F., Hua, Y., Qu, C.-B., et al. (2016), *Greatly enhanced cryogenic mechanical properties of short carbon fiber/polyethersulfone composites by graphene oxide coating*, Compos. Part A: Appl. Sci. Manuf. 89: 47-55. doi: 10.1016/j.compositesa.2016.02.016
7. Li, S., Chen, D., Yuan, Y., et al. (2020), *Influence of flexible molecular structure on the cryogenic mechanical properties of epoxy matrix and carbon fiber/epoxy composite laminate*, Mater. Des. 195: 109028. doi: 10.1016/j.matdes.2020.109028
8. Coronado, P., Argüelles, A., Viña, J., Viña, I. (2014), *Influence of low temperatures on the phenomenon of delamination of mode I fracture in carbon-fibre/epoxy composites under fatigue loading*, Compos. Struct. 112: 188-193. doi: 10.1016/j.compstruct.2014.02.007
9. Jia, Z., Li, T., Chiang, F.-P., Wang, L. (2018), *An experimental investigation of the temperature effect on the mechanics of carbon fiber reinforced polymer composites*, Compos. Sci. Technol. 154: 53-63. doi: 10.1016/j.compscitech.2017.11.015
10. Elmiladi, A.A., Petrović, V., Grbović, A., et al. (2021), *Numerical evaluation of thermal stresses generated in laminated composite structure exposed to low temperatures*, Therm. Sci. 25 (5B): 3847-3856. doi: 10.2298/TSCI200729338E
11. Nguyen, L.T.H., Hwang, J.-S., Kim, M.-S., et al. (2019), *Charpy impact properties of hydrogen-exposed 316L stainless steel at ambient and cryogenic temperatures*, Metals, 9(6): 625. doi: 10.3390/met9060625
12. Bartolomé, E., Bozzo, B., Sevilla, P., et al. (2017), *ABS 3D printed solutions for cryogenic applications*, Cryogenics, 82: 30-37. doi: 10.1016/j.cryogenics.2017.01.005

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