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COMPOSITES THEORY AND PRACTICE

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THERMAL ANALYSIS OF 3D-CF/Al-MMC BY MEANS OF DSC AND DILATOMETRY TESTS

Increasing demands for energy efficient lightweight structures capable of working in complex termomechanical loading conditions require the selection of appropriate load adapted materials as well as the development of associated manufacture processes ready for series production. Metal matrix composites with their advantages such as high thermal stability, reinforcement structure designability and high specific material properties have become more popular among materials applied as reliable load bearing products. To adapt the manufacturing process for these composites, understanding of solidification processes, the type and formation of interface and thermal behaviour of the composite are indispensable. The aim of this work was to understand the thermal behaviour of aluminium based metal matrix composites reinforced with carbon fibres 3D textile manufactured via the high pressure die casting (HPDC) process. Within the residual stress analysis and the sensitivity analysis of fibre coatings, plane specimens made of Al-226D with a carbon fibre (HTS) reinforcement with a nickel phosphorus coating have been investigated. Differential scanning calorimetry (DSC) and dilatometry tests have been performed considering the anisotropy of textiles and the formation of a crystallisation front in the cast specimens.

Keywords: thermal analysis, metal matrix composites, aluminium alloys, carbon fibre

ANALIZA TERMICZNA KOMPOZYTÓW 3D-CF/Al-MMC ZA POMOCĄ DSC ORAZ DYLATOMETRII

Wzrastający popyt na kompozyty na osnowie metalowej charakteryzujące się zdolnością do pracy w złożonych warunkach termomechanicznych wymaga dokładnej selekcji materiałów, jak również stworzenia dostosowanego procesu produkcji seryjnej. Kompozyty o osnowie metali lekkich posiadają szereg zalet, dzięki którym stają się coraz bardziej popularne wśród materiałów przeznaczanych na odpowiedzialne konstrukcje. Z tych zalet między innymi należy wyróżnić: wysoką stabilność temperaturową, elastyczność w projektowaniu struktury umocnienia oraz wysokie własności mechaniczne w odniesieniu do gęstości. W celu zoptymalizowania i adaptacji procesu produkcji kompozytów do warunków wielkoseryjnych niezbędne jest zrozumienie własności termicznych wytworzonego materiału. Celem tej pracy było wyznaczenie podstawowych własności termicznych kompozytu na osnowie stopu aluminium wzmacnianego włóknem węglowym w postaci trójwymiarowych tkanin, w procesie odlewania wysokociśnieniowego w kokilach. Włókno węglowe T800 infiltrowane było przez stop AlSi9Cu3(Fe) o numerze normy 226D. Z uwagi na bardzo dużą rozbieżność współczynników rozszerzalności cieplnej osnowy i wzmocnienia granica tych dwóch faz jest narażona na powstawanie naprężeń termicznych. Aby zniwelować powstałe naprężenia, niezbędne jest stosowanie pokrycia włókien węglowych przed procesem infiltracji. W pracy przeprowadzono badania skaningowej kalorymetrii różnicowej (DSC) oraz dylatometrii. Wykonano próby na próbkach nieumocnionych, umocnionych włóknami węglowymi oraz umocnionych włóknami węglowymi uprzednio pokrytymi powłoką niklowo-fosforową (NiP). Praca ta ukazuje, iż pokrywanie włókien węglowych powłoką na bazie niklu znacznie zmniejsza podatność produktu na mikropęknięcia, jak również ilość naprężeń termicznych powstałych w procesie produkcji odlewniczymi metodami wielkoseryjnymi.

Słowa kluczowe: analiza termiczna, kompozyty metalowe, stopy aluminium, włókno węglowe

INTRODUCTION

Metal matrix composites have become more popular due to the increasing demand for material characterised by high-strength, light-weight and high thermal stability. Weight reduction by applying advanced materials into product designed for use in complex thermomechanical loading conditions has drawn significant attention of the aerospace and automotive industry.

Aluminium metal matrix composites reinforced with carbon fibre 3D fabric (CF 3D/Al-MMC) are good examples of material combinations that lead to an increase in the lightweight potential. The main obstacle in manufacturing composites reinforced with carbon fibre is the susceptibility to form aluminium carbides in Al-based matrix alloys. These carbides occur at the interface of both phases, increase the brittleness of the composite and contribute to microcracks formation. Manufactured by the high pressure die casting (HPDC) method (with low time of contact between carbon fibre and liquid aluminium alloy), aluminium matrix composites reinforced with carbon fibre are characterised by good interface bonding with a lower amount of carbides. However, the HPDC method can contribute to the presence of thermal stresses, caused by the mismatch of thermal properties between the reinforcement and matrix, and also by an extremely high cooling rate [1].

To reduce the thermal stresses, the relaxation of a manufactured composite is indispensable. However, thermal treatments by annealing can exhibit a disadvantageous influence on the mechanical properties of the composite and very often cause enlargement of microfractures and carbides [2]. Therefore it is extremely vital to set the proper relaxation temperature, which will reduce the internal stresses while simultaneously limiting the formation and growth of carbides.

The presence of manufactured micro-failures has been investigated by computer tomography and initial microscopic analysis to find the proper cutting area of specimens. To determine the thermal behaviour of composites, dylatometry and differential scanning calorimetry (DSC) have been done with three different types of plates: unreinforced, reinforced with carbon fibre 3D-textile and reinforced with a nickel phosphorus (NiP) coated carbon fibre 3D-textile [3].

PRINCIPLES OF MANUFACTURING PROCESS

The manufacturing of metal matrix composites designed for use in complex loading conditions requires finding the proper method that allows the composite to attain the desired properties. The structure of the composites and their properties directly depend on the type of further applications. Regarding the demands of the industry in which, inter alia, a large batch production rate is one of the most important aspects, high pressure die casting has been chosen [4, 5]. The selected HPDC method is characterized by high pressures and high injection velocity of molten alloy into the mould. Moreover, the high production rate and use of steel moulds urges one to follow the rapid releasing of cast plates from the mould, which contributes to the fast cooling rate of the produced composites. Due to these aspects and also to receive good infiltration, additional preheating of the carbon fibre by infrared lamps directly before the injection has been done. The composite casting process can be divided into five steps:

1. setting up of mould containing attached double layer carbon fibre reinforcement
2. heating up of reinforcement by infrared lamps
3. closing of the die
4. aluminium alloy injection
5. release of cast plate

QUALITY ANALYSIS

Regarding the high velocity of molten aluminium injection, the initial preheating of the reinforcement and fast cooling rate, quality analysis is indispensable by means of computer tomography to determine the places most exposed to porosity, fractures and to estimate the porosity rate, fracture rate and amount of carbide creating the composite. Moreover, by means of computer tomography, the arrangement of carbon fibre reinforcement after casting can be observed and analysed. It is shown that extremely high pressures of molten metal injection hamper the receiving of a proper arrangement of textile reinforcement in a cast plate.

The strength of the composite highly depends on the adhesion between the matrix and reinforcement. The quality of infiltration and microstructure of the aluminium alloy at the boundary with the carbon fibre bundles has been done by microscopic analysis with the help of SEM equipped with EDX.

From the computer tomography analysis, the plates are divided to two quality types depending on the undulation rate of reinforcement (Fig. 1).

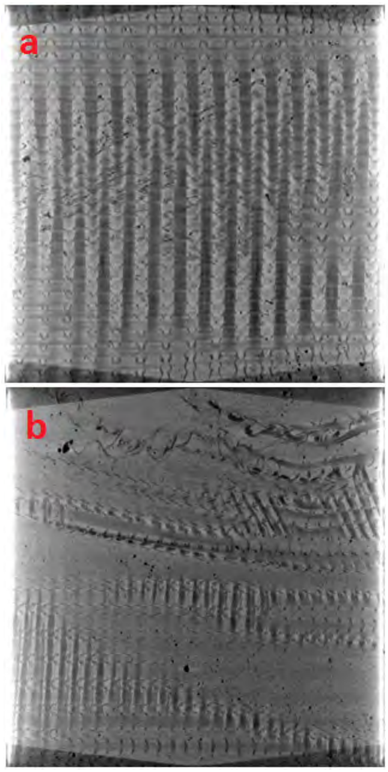

Fig. 1. Quality analysis of plates by CT: a) sufficient infiltration, b) insufficient infiltration

Rys. 1. Analiza jakości płyt za pomocą CT: a) wystarczająca jakość infiltracji, b) niewystarczająca jakość infiltracji

The microstructure of the specimens has shown a high amount of fractures at the contact surface between the infiltrated carbon fibres and the matrix for specimens reinforced with uncoated carbon fibre and a lower amount of fractures for specimens reinforced with Ni-coated carbon fibres (Fig. 2).

There are reasonable presumptions that the fractures are caused by the high cooling rate of the HPDC process and mismatch of thermal expansion coefficients of the aluminium matrix and carbon fibre reinforcement (Fig. 3) [6-8].

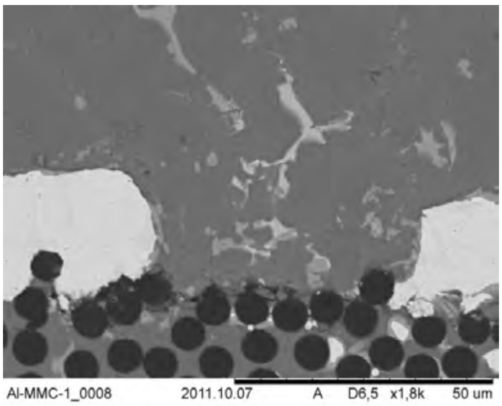

Fig. 2. Microstructure of 3D CF(NiP)/Al-MMC cast plate. Fracture occured at boundary between matrix and reinforcement without nickel-phosphorus coating

Rys. 2. Zdjęcie mikrostruktury płyty 3D CF(NiP)/Al-MMC. Widoczne pęknięcie występujące na granicy kontaktu pomiędzy wzmocnieniem a osnową w miejscu braku pokrycia niklowo-fosforowego

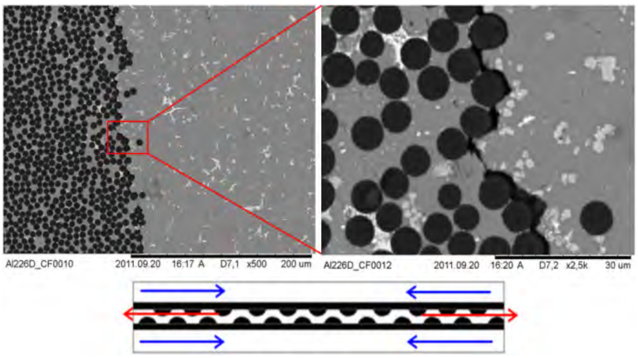

Fig. 3. Fractures at boundary between matrix and reinforcement of 3D CF/Al-MMC cast plate caused by mismatch of thermal expansion coefficient

Rys. 3. Pęknięcie na granicy osnowa-wzmocnienie w płycie 3D CF/Al-MMC spowodowane różnicą współczynników rozszerzalności cieplnej obu materiałów

SPECIMEN PREPARATION

The specimens prepared for DSC analysis have dimensions about 3 mm x 3 mm x 1 mm and have been cut from the middle and side part of a cast plate considering the crystallization front. The specimens for dilatometry analysis with dimensions about 3 mm x 3 mm x 20 mm have been cut from the middle part of the plate with two (0° and 90°) directions considering the reinforcement arrangement (Fig. 4).

The tests have been performed by a DSC (Mettler Toledo GmbH) and Dylatometry machine (LINSEIS Messgeräte GmbH) located at ILK, TU Dresden. The speed of the temperature rising for the DSC and Dylatometry test was set at about 10°C/min. The tests proceeded till 500°C.

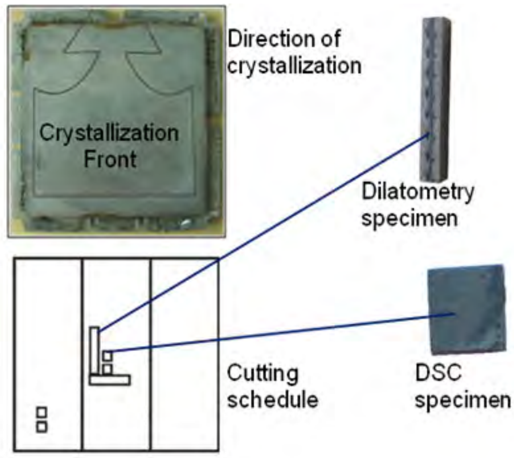

Fig. 4. Cutting lay-out of specimens

Rys. 4. Schemat cięcia próbek

PRINCIPLES OF THERMAL ANALYSIS

Differential Scanning Calorimetry

Differential Scanning Calorimetry (DSC) is used to determine physical conversions like glass transition temperatures, melting and crystallisation effects. Furthermore, chemical reactions like cross linking and curing reactions of thermosets and elastomers can be characterised. This method is also used to determine the specific heat capacity. The DSC measuring principle is based on the measurement of the caloric effects of a sample comparing to a reference substance. Therefore, the sample and reference substance are heated within one furnace at a defined heating rate.

In that way, the effective temperatures of the sample and the reference material are simultaneously measured. In regard to technical applications, the change of enthalpy ΔH is an important parameter. It is calculated as the integral of heat capacity c_p according to the measured temperature change (dT) as shown in the following equation

$$\Delta H = \int c_p \cdot dT \qquad (1)$$

In this context, the effects which cause a rise in the enthalpy; for example vaporisation, melting or glass transition are called endothermic processes. The effects reducing the enthalpy e.g. crystallisation, decomposition, and curing are called exothermic processes. Figure 5 shows a schematic drawing of the characteristic effects in a time or temperature dependent heat flow diagram.

Here the used DSC device is not able to measure the heat capacity directly. Therefore the heat flow is determined, which is characterized by the heat quantity per unit of time and weight m. This value is directly proportional to the specific heat capacity with heating rate v as a proportionality factor

$$\frac{\dot{Q}}{m} = v \cdot c_p \qquad (2)$$

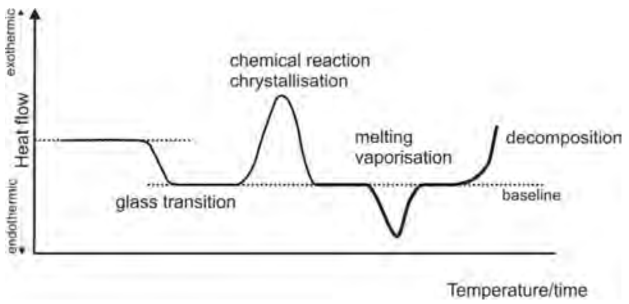

Fig. 5. Example of exo- and endothermic effects during DSC measurement [9]

Rys. 5. Przykładowe efekty egzo- i endotermiczne występujące podczas badania DSC [9]

For these investigations, a DSC (Mettler Toledo GmbH) has been used. There the integrated temperature sensor FRS 5 consists of 56 thermocouples for a high precision temperature measurement. Measurements in the temperature range of −100°C up to 700°C and heating rates up to 70 K/min as well as cooling rates up to 20 K/min are possible. In this range, the temperature accuracy is ±0.2 K and the sensitivity ±0.02 K. The maximum sample rate is 50 Hz and the measuring range of the heat flow ± 350 mW at a resolution of 0.04 μW.

Dilatometry

The materials show temperature dependent expansion behaviours. Therefore, dilatometry offers the possibility to precisely measure the dimension changes especially of solids but also for melts, powders and pastes according to a defined temperature change. The main application field of the dilatometry is the evaluation of sintering processes of ceramics, metals and powder metals. Furthermore, dimensional changes during the chemical reactions and phase changes of the materials can be detected. For characterization of the thermal expansion behaviour, the technical coefficient of expansion α_{tech} is of important relevance. It is calculated with initial length l_0 at reference temperature T_0 (in general 20°C), length l_1 at a second temperature T_1 by the following equation

$$\alpha_{tech} = \frac{1}{l_0} * \frac{l_1 - l_0}{T_1 - T_0} \tag{3}$$

In the ILK thermal laboratory, the dilatometer (Type L75) from LINSEIS Messgeräte GmbH with a vertical built up has been used. There the sample is vertically positioned in a protective conduit and clamped between a push rod and an upper contact leaf (Fig. 6). The pressure load (in general 100 mN up to 300 mN) for the clamping is controlled and constant throughout all the testing.

The length of the measuring device is thermally decoupled from the sample measuring system and constantly tempered. The dimension changes are detected by a LVDT sensor, which allows resolutions of 0.125 nm/digit. In general, the sample geometry should be cylindrical with dimensions of 5 mm in diameter and a length of 20 mm. However, samples with a square cross section are used as well. The temperature is measured and controlled by a type K thermocouple.

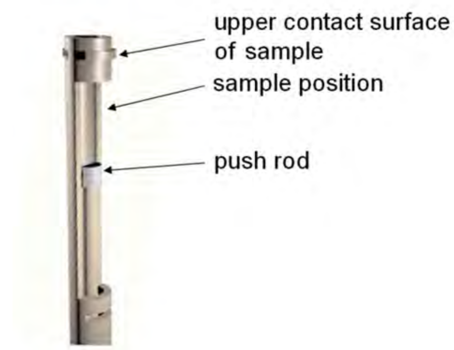

Fig. 6. Al_2O_3 measuring system standard (based on LINSEIS Messgeräte GmbH)

Rys. 6. Standardowy system pomiarowy - Al_2O_3 na podstawie Lineseis Messgeräte GmbH

RESULTS AND DISCUSSION

The dilatometry analysis of specimens cut out of an unreinforced plate has shown a rise in the thermal expansion coefficient of about 200÷250°C (Fig. 7).

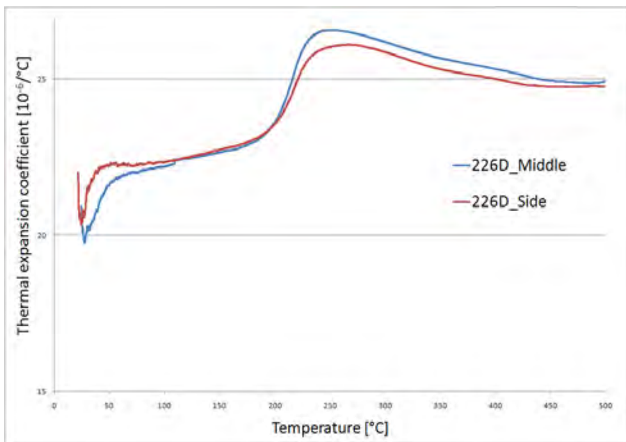

Fig. 7. Dilatometry results of Al-226D cast plate

Rys. 7. Wyniki dylatometrii dla płyty Al-226D

There were reasonable presumptions that the shape of the studied curves should be similar for all of the samples taken from the unreinforced part of the specimens. However, the dilatometric analysis of the specimens reinforced with uncoated carbon fibres has shown a curve that deviates from the expectations (Fig. 8). Although the reinforced part of specimens showed a lower increase in thermal expansion coefficient at the temperature of 200÷250°C, the matrix of these plates shows a lack of increase in the mentioned temperature range. It is probably caused by relaxation of the reinforced specimens by fracturing at the surface of the bundles.

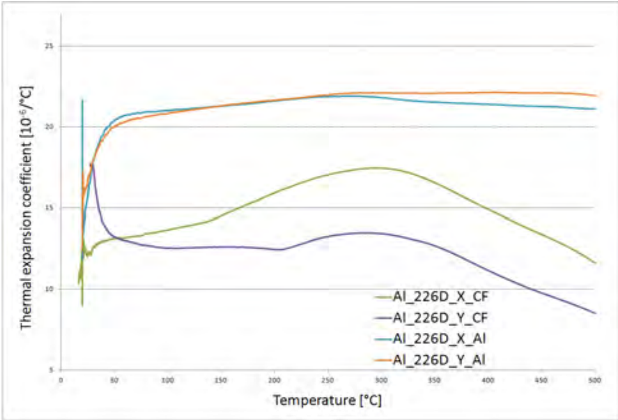

Fig. 8. Dilatometry results of 3D-CF/Al-MMC cast plate

Rys. 8. Wyniki dylatometrii dla płyty 3D-CF/Al-MMC

The Ni-coated specimens have shown a higher increase in TEC at the temperature range and also a lower exhibition of fractures at microscopic analysis (Fig. 9).

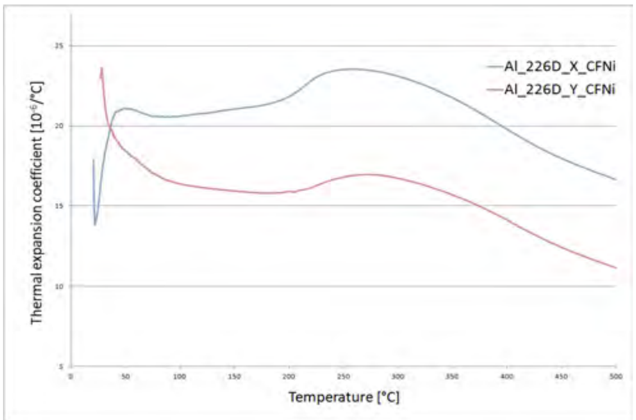

Fig. 9. Dilatometry results of 3D-CF(NiP)/Al-MMC cast plate

Rys. 9. Wyniki dylatometrii dla płyty 3D-CD(NiP)/Al-MMC

The dilatometry analysis of all the specimens show interesting changes in the temperature range of about 200 to 300°C. Therefore, the DSC analysis of specimens in the same temperature range and similar conditions has been undertaken.

The DSC analysis of specimens from the unreinforced plate has shown an energy peak in the same temperature range as the dilatometry test results (Fig. 10).

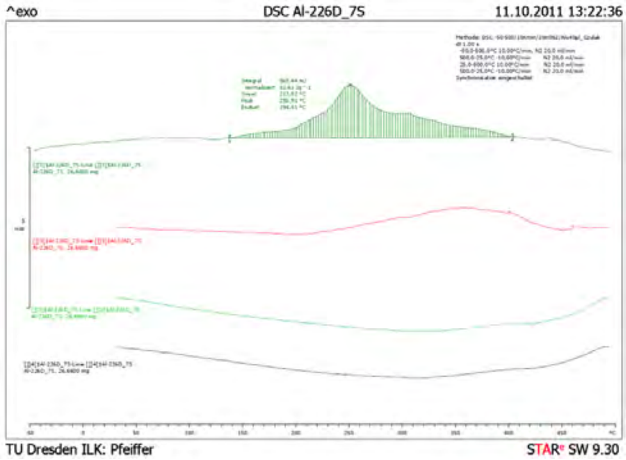

Fig. 10. DSC results of Al 226D cast plate

Rys. 10. Wyniki DSC dla płyty Al 226D

By analysing the carbon fibre reinforced specimens, a lack of peak for specimens from an unreinforced part of the plate is visible. This fact confirms the presumptions about the relaxation of specimens by fracturing. The DSC from the reinforced part of the plate has shown another interesting peak related to the infiltrated reinforcement in the specimen (Fig. 11).

The DSC analysis of the specimens from the plate reinforced with NiP-coated carbon fibres (Fig. 12) has shown a normal energy peak of the matrix (similar to Al 226D results) and a flat characteristic of curve for the reinforced part of the specimens (similar to results from reinforced part of 3D CF/Al-MMC).

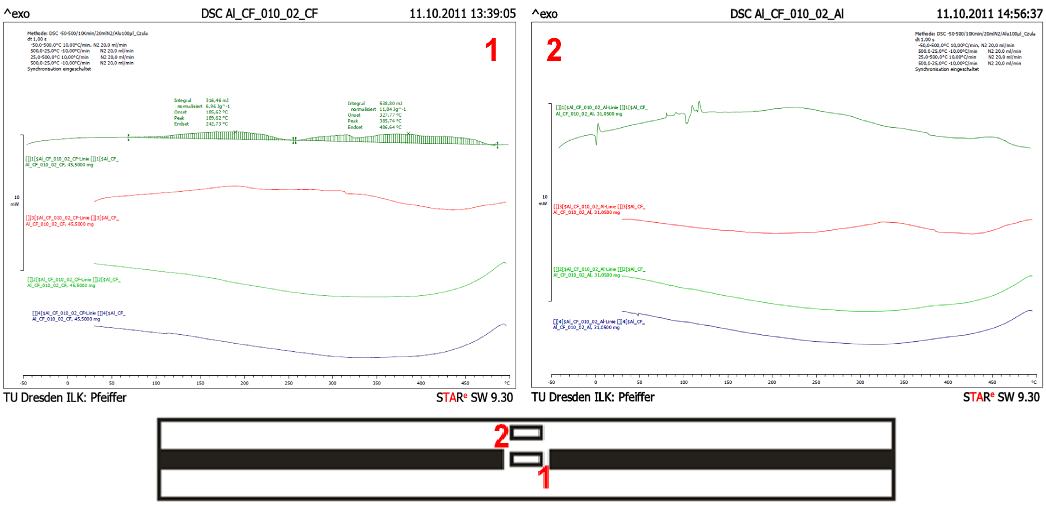

Fig. 11. DSC results of 3D CF/Al-MMC cast plate

Rys. 11. Wyniki DSC dla płyty 3D CF/Al-MMC

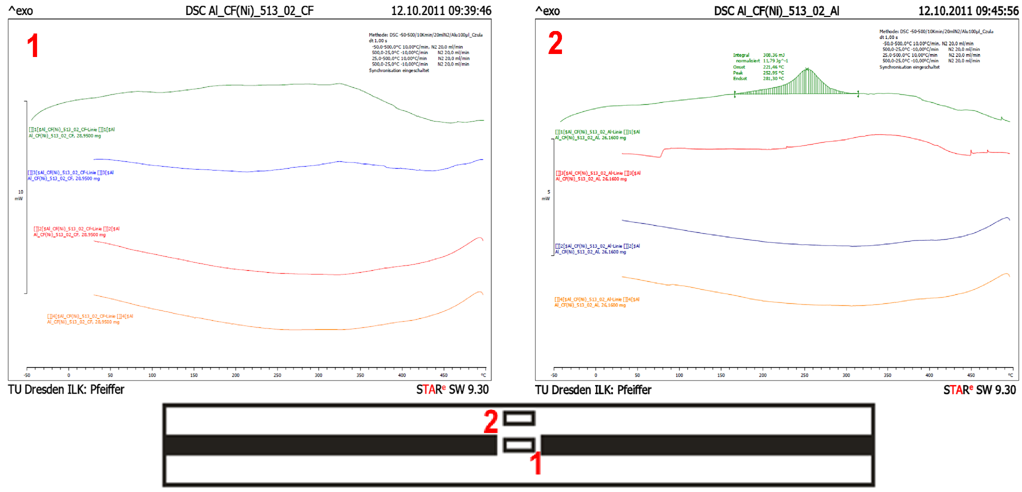

Fig. 12. DSC results of 3D CF(NiP)/Al-MMC cast plate

Rys. 12. Wyniki DSC dla płyty 3D CF(NiP)/Al-MMC

This fact shows the advantageous influence of NiP coating on the thermal behaviour of the CF/Al composite by reducing the fracturing effects and creation of thermal stresses.

SUMMARY

Thermal analysis of the carbon fibre 3D-textile reinforced aluminium alloy manufactured by the HPDC process has shown various changes in the thermal behaviour of the composite. Unreinforced plates cast with the 226D aluminium alloy show interesting changes of the TEC in the temperature range of 200÷250°C. Similar changes have been shown by the DSC analysis of this plate, where the curve of the first heating shows an exothermal energy peak in the temperature range of about 150÷400°C in which the maximum of the peak was always placed at 250°C.

It is assumed that the thermal analysis results are related to the relaxation of the specimens. However, the plate reinforced with the uncoated carbon textile has shown different results of thermal behaviour. The lack of energy peak of DSC analysis of the unreinforced part of the CF/Al-MMC plate can be caused by internal fracturing of the specimens. Interfacial fracturing is caused by a mismatch of thermal expansion coefficients and extremely fast cooling rate. The results have shown that using an NiP coating is advantageous for the thermal behaviour of CF/Al-MMC composites. The specimens from the unreinforced part of Ni-coated textile reinforced specimens show a lack of fractures in the vicinity of the cast areas.

Thermal analysis with computer tomography can give valuable and interesting information about thermal residual stresses in composite materials. However, to investigate the thermal behaviour of composites based on an aluminium matrix and coated carbon fibres, further DSC and dilatometry investigations are necessary.

REFERENCES

[1] Giuliani A., Albertini G., Manescu A., Residual stress analysis on tensile MMC specimens after loading/unloading tests in several conditions, Physica B 2004, 350.

[2] Yang M., Scott V.D., Carbide formation in a carbon fibre reinforced aluminium composite, Carbon 1991, 29, 7, 877--879.

[3] Bruno G., Ceretti M., Girardin E., Manescu A., Relaxation of residual stress in MMC after combined plastic deformation and heat treatment, Scripta Materialia 2004, 51.

[4] Rübner M., Günzl M., Körner C., Singer R.F., Aluminium--aluminium compound fabrication by high pressure die casting, Materials Science and Engineering A 2011, August, 528, 22-23, 25, 7024-7029.

[5] Qianqian Li, Rottmair Ch.A., Singer R.F., CNT reinforced light metal composites produced by melt stirring and by high pressure die casting, Composites Science and Technology 2010, December, 70, 16, 31, 2242-2247.

[6] Aghdam M.M., Khojeh A., More on the effects of thermal residual and hydrostatic stresses on yielding behavior of unidirectional composites, Composite Structures 2003, 62, 3-4.

[7] Ghonem H., Wen Y., Zheng D., An interactive simulation technique to determine the internal stress states in fiber reinforced metal matrix composites, Materials Science and Engineering A 1994, April, 177, 1-2, 15, 125-134.

[8] Cholewa M., Szuter T., Structure of AlSi skeleton castings, Archives of Foundry Engineering 2012, 12/2, 147-152.

[9] Ehrenstein G.W., Riedel G., Trawiel P., Praxis der thermischen Analyse von Kunststoffen 2003, Hanser Verlag (p. 1).

