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(54) Title: EDGE EFFECT WEAKENING BY MEANS OF BYPASS-CONDUCTOR DURING INDUCTION WELDING PROCESS

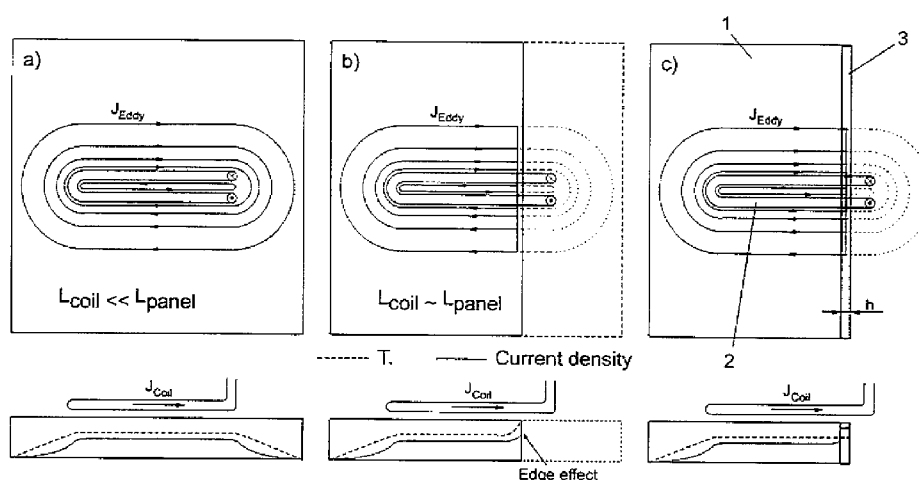


Fig. 2a-c

(57) Abstract: Method for induction welding of at least one panel (1) of electrically semi-conducting material with a specific mass of < 2 kg/l, comprising providing the at least one panel (1), providing an electrically conducting element (3) at at least one edge of the panel and applying an electro-magnetic field at the edge zone induction welding the edge zone. The presence of the conducting element (3) avoids the edge overheating during welding.



Title Edge effect weakening by means of bypass-conductor during induction welding process

FIELD OF THE INVENTION

5 The invention is in the field of an induction welding process particularly suited for thermoplastic composites.

BACKGROUND OF THE INVENTION

The invention is in the field of an induction welding process particularly suited for thermoplastic composites. 10 These composites can in principle be processed rapidly by thermoforming, i.e. forming under a heat source. The composites may therefore be applied in various applications, such as aerospace and automotive industry. Unfortunately the composites have not been applied significantly yet as the technology for joining is considered insufficient, especially for 15 pre-fabricated parts. It is considered that for thermoset composites complex pre-treatment and heating mechanisms are required, whereas for thermoplastic composites it is difficult to ensure that only the interface is heated and no (foreign) material is left at the bond line. Besides an esthetical effect the bonds have limited strength as a consequence and thus 20 limited application.

Thermosets composites have limited application as they are joined using mechanical fastening or adhesive bonding. For 25 mechanical fastening the holes involved cut reinforcement fibres and result in stress concentrations around the holes. Adhesive bonding may require large prefabricated parts to be heated, in order to cure the adhesive.

Thermoplastic composites can be joined by other techniques such as fusion or welding; this process does in principle not damage the composites and provides further advantages. For the sake of understanding a welding process of a thermoplastic composite is considered to relate to a process that heats the interface/surface to a temperature above a glass 35 transition temperature of the composite for amorphous polymers or above a melting point for semi-crystalline polymers. The process typically involves application of pressure such as to diffuse thermoplastic polymers over the interface/surface and thereby forming a welded joint, typically after cooling. The

process is typically fast without interface/surface preparation. Despite advantages there has not yet been much use of thermoplastic composites for applications, amongst others because a lack of joining technology.

5 Joining may be required when components of a product, such as an aircraft, are too large to be produced as a single part, when different materials are used, when different production conditions are needed, when components are made in physical locations spaced apart, etc.

10 Various approaches have been investigated for joining, such as mechanical fastening, fusion bonding, and adhesive bonding. As mentioned these bonding techniques suffer from various drawbacks, especially for thermoset materials. Mechanical fastening involves holes; by making holes load bearing fibers are
15 inevitably cut and stress around the holes is introduced. Adhesive bonding may require a full part to be heated in order to cure adhesive at the relatively small joint. Of the fusion bonding techniques being available induction fusion, resistive implant fusion and ultrasonic fusion are considered suitable.
20 For induction welding it is considered that heat is generated through heat losses from the induced eddy currents in response to an applied alternating magnetic field. In an approach induced eddy currents may flow in a metal implant which needs to be provided such as at a weld interface or in fibers of the
25 composite material. Induction welding processes have been further developed recently, in particular for airplanes. Considering random distributed fibers, which is typically the case, so-called volumetric heating may be achieved as closed electro-magnetic pathways in the composite material in the material itself are then provided by the fibers. However, on an
30 interface, such as between two surfaces, extra measures need to be taken, also in order to concentrate heat generated by induction, especially when thicker elements (> 4 mm) are concerned. Typically also subsequent panels/interfaces are rotated with respect to one and another, e.g. when forming a
35 laminate.

Some publications recite metallic samples, which are considered not well suited for electrically non-conducting or

semi-conducting materials. Nemkov et al. in "Simulation of induction heating of slabs using ELTA 6.0", Intern. Scientific Colloquium Modelling for Electromagnetic Processing, Hannover (2014) provide a modelling tool to optimize an induced power profile by changing a frequency of the induction welding, which affects the penetration depth. The method is limited to metallic samples. In addition low frequencies are required to achieve a sufficient penetration depth; however lower frequencies reduce the efficiency of the heating process. Forzan et al. in "Compensation of induction heating load edge-effect by space control", COMPEL-The international journal for computation and mathematics in electrical and electronic engineering, 30(5), 1558-1569 (2011) present a numerical method to optimize an induced power profile by connecting different coils along a sample. The method is limited to metallic samples. In addition a complex set-up is required with multiple coils and the method can only be used for simple geometries.

Some patent documents recite methods for induction welding.

US 5,500,511 A recites tailored susceptors for induction welding of thermoplastic, to obtain more uniform heating across the susceptor when welding composite parts using induction heating, having edge regions of lower absolute impedance or a lower longitudinal impedance than the transverse impedance to counterbalance the higher current density and current that occurs near the edges. The lower impedance at the edges is achieved by altering the aspect ratio (length/width) of openings in the susceptor, by folding the susceptor over onto itself, or both. Uniform heating is important to obtaining a uniform, consistent weld on which aerospace designers can rely.

US 5,508496 A recites salvaged susceptor for thermoplastic welding by induction heating which are tailored to provide precise control of the heat applied to the weld and especially to control overheating at the edges of susceptor. The preferred susceptor has a central portion having a uniform pattern of substantially similar openings and selvage edge strips devoid of openings to provide a low impedance current path for eddy currents at the edges. We can create aerospace structure, particularly a wing skin-spar assembly, without fasteners.

These patents are related to a susceptor which is provided as a metal mesh embedded inside the composite. The material is heat up by the metal mesh, rather than heating up the composite/carbon fiber itself. However, adding the mesh is found not sufficient to avoid the edge effect, which is affecting the metal mesh rather than the composite. They alleviate the effect by changing the aspect ratio of the mesh near the edges. But the addition of an external element "metal mesh" inside the composite increases the weight of a final part and decreases the strength of the joint and is thus not applicable in many cases.

US 6,939,477 A recites temperature-controlled induction heating of polymeric materials. Induction heating technologies are utilized to weld, forge, bond or set polymer materials. The invention provides controlled-temperature induction heating of polymeric materials by mixing ferromagnetic particles in the polymer to be heated. Temperature control is obtained by selecting ferromagnetic particles with a specific Curie temperature. The ferromagnetic particles will heat up in an induction field, through hysteresis losses, until they reach their Curie temperature. At that point, heat generation through hysteresis loss ceases. This invention is applicable to bonding thermoplastic materials, wherein only the area to be heated has ferromagnetic particles in it; bonding of thermoset composites, which have been processed with a layer of thermoplastic material on one side; curing of thermoset adhesives or composite resins; or consolidating thermoplastic composites.

US 5,248,864 A recites a method for heating a composite comprising a nonconductive material that is a thermoplastic or a thermoset reinforced with conductive materials such as carbon fibers, by incorporating coupling particles in the structure. The structure is oriented in the plane of a magnetic field induced at frequencies from 3 kHz to 7 MHz, whereby the coupling particles respond as susceptors to the induced magnetic field and are preferentially heated (carbon fibers are not heated).

These patents recite a susceptor in the form of metal particles mixed in the composite. The approach is to heat up the

material by such particles rather than the carbon fiber itself. It does not relate to a practical application as a high mass percentage of particles is required in order to be efficient; the particles affect the material properties of composite adversely. Also dispersing the particles in the matrix is a drawback.

US 2013/149,501 A1 recites a method for connecting a fibre composite component to a structural component of an aircraft and spacecraft. A metal foil is provided as a transverse reinforcement element between the fibre composite component and the structural component. It is formed with at least one anchoring portion which protrudes from the surface facing the fibre composite component and is inserted between the fibre composite component and the structural component. A corresponding arrangement is produced in accordance with this method.

WO 90/008027 A1 recites a method of joining plastics materials by forming a contact between two parts to be joined, placing an electrical induction coil adjacent the contact and electrically inductively heating the parts in the vicinity of the contact. A layer comprising multidirectional discontinuous carbon fibres in the presence of thermoplastics material is provided in the vicinity of the contact, at least some of the carbon fibers being oriented to provide resistive conducting paths for the flow of eddy currents. Electrically inducing eddy currents in the layer heats the thermoplastics material and causes it to fuse at the contact and form a join between the two parts.

The above two documents still suffer from edge overheating during welding.

The present invention therefore relates to an improved method for induction welding of panels, a construction element obtainable by said method, and a product obtainable by said method, which overcome one or more of the above disadvantages, without jeopardizing functionality and advantages.

SUMMARY OF THE INVENTION

The present invention relates in a first aspect to a method for induction welding of at least two panels. Said panels typically form part of a larger construction to be formed.

The panels are typically flat and relatively thin. The panels are typically limited in size, e.g. from a few cm by a few cm (e.g. 10 by 10 cm²) to a size that can still be handled, either by a machine or by a human being (e.g. 10 by 10 m²). The present panels are formed of electrically semi-conducting materials, which materials are well suited for induction welding. Also the present panels have a relatively low specific mass of < 2 kg/l, preferably of < 1.5 kg/l, more preferably of < 1.4 kg/l, such as < 1.35 kg/l, especially when compared to prior art construction materials, such as aluminium or (light weight) metal in general. In view of displacement of a final product such a low specific mass and hence low final mass is highly desired. First at least one panel is provided. The panel may be provided on an intermediate, i.e. non-final, product; on said intermediate product an earlier applied panel may already be present, said earlier applied panel having an edge (as well). The at least one panel has at least one edge, i.e. a virtual line where the panel physically ends. On at least one edge of the panel an electrically conducting element is provided, typically leaving a distance open, measured (perpendicular) from the edge, of less than 5 cm, preferably less than 1 cm, more preferably less than 5 mm, even more preferably less than 1 mm, such as < 0.1 mm or about 0 cm; in fact the conducting element may be partly over the edge, such as 0.1-1 mm. Said conducting element, which may also be referred to as "by-pass conductor", is found to provide improved characteristics at the edge zone and mitigates the above mentioned problems. The conducting element preferably has a good electrical contact with the panel. Then an electro-magnetic field at an edge zone of said panel comprising said electrically conducting element is generated, and thereby the edge zone is induction welded. Therewith strong joints have been obtained of e.g. > 40 MPa inter laminar shear strength (in average), e.g. using a Zwick Roell according to EN 2563, as well as low porosities. In addition the temperature distribution during welding is homogenous in the join, thereby optimizing the quality of weld. As mentioned, no external insert (susceptor) is needed, thus the overall weight of the composite is reduced.

Material optimization by reducing the overlap area to be weld, which turns to a reduction of weight and material costs.

The invention provides a novel methodology to overcome undesired edge effects when two light materials (such as based on composites of carbon fibers and plastics) are weld by induction welding. The challenge is found to be to properly heat up an panel edge zone to weld at a homogenous temperature. The disadvantageous edge effect occurs when a coil is near the edge, concentrating electrical currents which overheat the panel and sometime burn spots can even appear. A prior art solution is to avoid welding near the edge, thereby reducing material optimization and control of a target temperature profile. It is noted that underling physics of induction welding is not well understood yet. A mechanism is not obvious even for experts and could e.g. be misunderstood to relate to a heat element rather than to an electrical element.

Suitable materials for panels and final products typically have a Young's modulus (E) of > 1 GPa, such as > 3 GPa, a tensile strength of > 50 MPa (ISO 527), such as > 90 MPa, an elongation @ break of > 40% (ISO 37:2011), such as > 50%, a glass transition temperature of > 100 °C (as determined by differential scanning calorimetry), such as > 130 °C, a melting point of > 250 °C, such as > 300 °C, a thermal conductivity of 0.1-1 W/m.K, and a low water absorption (24 hours (ASTM D 570) < 0.1%), and combinations thereof.

In a second aspect the present invention relates to a construction element obtainable by the present method, wherein a porosity percentage of the element at the edge zone is < 7%, preferably <3%, more preferably <2%, such as <1% and typically <0.5%, as determined by ultrasound or computed radiography. Such a porosity can be determined according to the procedure as described in e.g. J. Composites, Vol. 2013, Article ID 140127, "Porosity Distribution in Composite Structures with Infrared Thermography" by C. Toscano et al.

In a third aspect the present invention relates to a product obtainable by the present method, wherein the product is selected from a part of transport vehicle or the vehicle, such as an airplane, a car, a truck, a boat, a ship, wherein the part is preferably one of a wing, a tail, a door, a body, a

door, a hood, and a frame.

Thereby the present invention provides a solution to one or more of the above-mentioned problems.

Advantages of the present invention are detailed throughout
5 the description.

DETAILED DESCRIPTION OF THE INVENTION

The present invention relates in a first aspect to a method according to claim 1.

In an exemplary embodiment of the present method at
10 least two panels are provided of different materials or of the same material, with the proviso that the to be welded at least one panel is typically of a thermoplastic material. In this respect a panel may also relate to an intermediate product comprising e.g. an earlier applied panel having an
15 edge, on which intermediate product a further panel is applied, typically edge to edge. The intermediate product and/or earlier applied panel may be of the same material as the at least one panel applied subsequently or of a different material.

20 In an exemplary embodiment of the present method the material(s) is/are a composite material, such as a thermoplastic composite or a thermoset composite, such as a carbon or glass reinforced material, and combinations thereof.

In an exemplary embodiment of the present method the material or materials, and especially the to be welded panel,
25 has/have an electrical conductivity of $< 10^4$ S/m at 20 °C, i.e. are slightly conducting. In order to provide good welding properties, and optionally to alleviate electrical fields, such as those generated by friction/resistance, some
30 electrical conductivity is preferred.

In an exemplary embodiment of the present method the material(s) is/are selected from aromatic or aliphatic polymers, such as polyether ketones, such as polyaryletherketones, such as Polyether-ether-ketone (PEEK), Polyether-ketone-ketone
35 (PEKK), poly sulfones, such as poly-ether sulfones (PES), and poly phenyl sulfones (PPSU), poly sulphides, such as poly phenylene sulphide (PPS), poly alkylenes, such as poly alkylene imines, such as poly alkylene imines, such as poly

ethylene imine (PEI), poly imidazoles, such as polybenzimidazole (PBI), and combinations thereof, preferably PEI, PES, PEEK, PEKK, PPS and PEI.

In an exemplary embodiment of the present method a
5 thickness of the at least one panel is from 0.2-20 mm, preferably 0.5-5 mm, more preferably 0.6-3 mm, even more preferably 0.7-2 mm, such as 1-1.5, hence relatively thin panels may be welded.

In an exemplary embodiment of the present method the
10 conducting element is selected from a conducting tape, such as a copper tape, a conducting grease, such as copper or aluminum grease, graphite, graphene, a coating, such as a metallic coating, such as a gold coating.

In an exemplary embodiment of the present method the
15 conducting element has a thickness (t) of 10-1000 μm , preferably 20-500 μm , more preferably 30-200 μm , such as 50-100 μm , e.g. 70-80 μm , and/or a relative width of 50-100% of the thickness of the panel, preferably 70-99% of said thickness, more preferably 90-98% of said thickness. It has been found
20 experimentally that e.g. in view of a temperature profile obtained during induction welding, the width of the conducting element is not too large and not too small.

In an exemplary embodiment of the present method the
conducting element has an electrical conductivity of $> 10^3$
25 S/m at 20 °C, preferably $> 10^4$ S/m, more preferably $> 10^6$ S/m, i.e. a relatively high conductivity. It has been found that the higher the conductivity is, the better results are obtained and less conducting material needs to be used.

In an exemplary embodiment of the present method the
30 conducting element is provided over a width of 1-1000 cm, preferably 2-500 cm, such as 5-250 cm.

In an exemplary embodiment of the present method the
edge is polished before applying the conducting element, such
as by sand polishing, preferably using a fine polishing mate-
35 rial. In addition the edge may be cleaned, such as chemically cleaned, in order to remove impurities. Especially polishing is found to reduce adverse porosity effects.

In an exemplary embodiment of the present method a welding temperature is from 423-673 K (150-400 °C), preferably

448-623 K (175-325 °C), more preferably 473-573 K (200-300 °C), and/or at a magnetic field density of 10-50 Gauss, such as 20-30 Gauss.

In an exemplary embodiment of the present method a frequency of 0.01-10 MHz is used, preferably 0.1-5 MHz, such as 0.15-3 MHz, and/or a power of 10-1000 W/cm², preferably 20-500 W/cm², more preferably 50-250 W/cm², and/or during a period of 1-120 sec, preferably 2-60 sec, and/or at a relative welding speed of 5-200 mm/sec, preferably 10-100 mm/sec, such as 15-50 mm/sec. Said welding speed may be obtained by moving the panel relative to the welding element, by moving the welding element, or both

In an exemplary embodiment of the present method the panels form a part of transport vehicle, such as an airplane, a car, a truck, a boat, a ship, wherein the part is preferably one of a wing, a tail, a door, a body, a door, a hood, and a frame.

The invention is further detailed by the accompanying figures and examples, which are exemplary and explanatory of nature and are not limiting the scope of the invention. To the person skilled in the art it may be clear that many variants, being obvious or not, may be conceivable falling within the scope of protection, defined by the present claims.

SUMMARAY OF THE FIGURES

Figure 1. Photographs of various panels

Figure 2a-c. Eddy current distribution.

Figure 3. Temperature profile in cross section.

DETAILED DESCRIPTION OF THE FIGURES

Fig. 1. Photographs of various panels, the top panel being a carbon fiber composite without conducting element, there beneath an aluminum grease applied to the edge, there beneath a gold coating applied, and the bottom panel with a reversed copper tape.

Fig.2a-c show schematically the eddy current distribution for a coil being much smaller than the panel (2a), for a coil being of similar dimensions as the panel (2b), and the effect of the present conducting element (2c). In figure 2 edge ef-

fect strength is given in view of relative dimensions of composite panel (1) and coil (2). Dimensions: a) Panel dimensions much larger than the coil ($L_{coil} \ll L_{panel}$); b) Panel dimensions similar or shorter than the coil ($L_{coil} \sim L_{panel}$), inducing high current density near the edge; c) Mechanism of bypassing the current density through the thickness (h) of the bypass-conductor (3).

Fig. 3 shows an exemplary effect of the thickness h of the applied conducting element. The temperature profile (T in °C on the vertical axis) is given as a function of position on the panel (horizontal axis, in mm). It can be observed that the thickness of the conducting material is especially of influence on the temperature near the edge, e.g. the last 10-20 mm edge zone. A plot of the temperature profile of the composite (1) along the panel length (L) in cross section (1.a) for different thicknesses (h) by using copper as a bypass conductor (2). The optimal range is found to be between 60µm and 100µm in thickness (red curves).

The figures are further detailed in the description and examples below.

EXAMPLES/EXPERIMENTS

The below relates to an example of the present induction welding. The induction welding is carried out according to the following steps: First a composite is placed near the coil with a gap called coupling distance (typically $CD = 3-6$ mm). Then, a consolidation pressure is applied on the join of >6 bars (which may be up to 10 bars). Next, the induction welding equipment is switched on with settings of currents and time (a frequency is typically not controlled). Finally the coil is slid along the join keeping the same coupling distance.

All the test are carried out with the same alternating current of coil IRMS = 100A during 100 seconds and the same coupling distance ($CD=5$ mm) with a resonance frequency of 375 Khz. The setup is designed in such a way that is not affecting the magnetic field. A static support is made by wood where the corners of the panel are resting on, in order to minimize the heat transfer by contact. Hence, the wood support is not taken into account in the model. The hairpin coil is placed centred

under the panel with the dimensions as sketched in the figures. The gap between the copper coil and the bottom surface of the panel is known as coupling distance which is 5mm (CD=5mm). The temperature is recorded using a regular thermocouple transducer type K, however the metallic tip can be effected by induced currents in the vicinity of the coil. Hence, it is place on the top surface of the panel because the magnetic field strength is considered to decrease through the thickness of the composite and at the centre where the magnetic field is weaker due to the coil's shape, reducing the effect that the magnetic field could produce to the sensor. The thermocouple is also electrically isolated from the composite by placing Kapton tape between them, preventing any parasite current flowing through. A thermal infrared camera from Flir Systems type A615 is set 40 cm above the panel to capture the heat pattern on top surface. Both devices are recording each second of all the data which is stored in a PC. As such a good adhesion is obtained.

CLAIMS

1. Method for induction welding of at least one panel of electrically semi-conducting material with a specific mass of < 2 kg/l, comprising

5 providing the at least one panel, the panel having at least one edge,
providing an electrically conducting element at at least one edge of the at least one panel,
generating an electro-magnetic field at an edge zone of said panel comprising said electrically conducting element,
10 and
induction welding the edge zone.

2. Method according to claim 1, wherein at least two panels are provided of different materials or of the same material.

3. Method according to any of the preceding claims, wherein
15 the material(s) is/are a composite material, such as a thermo-plastic composite or a thermoset composite, such as a carbon or glass reinforced material. Combination?

4. Method according to any of the preceding claims, wherein
20 the material or materials has/have an electrical conductivity of < 10^4 S/m at 20 °C.

5. Method according to any of the preceding claims, wherein the material(s) is/are selected from aromatic or aliphatic polymers, such as polyether ketones, such as polyarylether-
ketones, such as Polyether-ether-ketone (PEEK), Polyether-ke-
25 tone-ketone (PEKK), poly sulfones, such as poly-ether sul- fones (PES), and poly phenyl sulfones (PPSU), poly sul- phides, such as poly phenylene sulphide (PPS), poly al- kylenes, such as poly alkylene imines, such as poly alkylene imines, such as poly ethylene imine (PEI), poly imidazoles,
30 such as polybenzimidazole (PBI), and combinations thereof.

6. Method according to any of the preceding claims, wherein a thickness of the at least one panel is from 0.2-20 mm, preferably 0.5-5 mm.

7. Method according to any of the preceding claims, wherein
35 the conducting element is selected from a conducting tape, such as a copper tape, a conducting grease, graphite, gra- phene, a coating, such as a metallic coating, such as a gold

coating.

8. Method according to any of the preceding claims, wherein the conducting element has a thickness (t) of 10-1000 μm , preferably 20-500 μm , more preferably 30-200 μm , such as 50-
5 100 μm , e.g. 70-80 μm , and/or a relative width of 50-100% of the thickness of the panel, preferably 70-99% of said thickness, more preferably 90-98% of said thickness.

9. Method according to any of the preceding claims, wherein the conducting element has an electrical conductivity of $> 10^3$
10 S/m at 20 °C, preferably $> 10^4$ S/m, more preferably $> 10^6$ S/m.

10. Method according to any of the preceding claims, wherein the conducting element is provided over a width of 1-1000 cm.

11. Method according to any of the preceding claims,
15 wherein the edge is polished before applying the conducting element.

12. Method according to any of the preceding claims, wherein a welding temperature is from 423-673 K (150-400 °C), preferably 448-623 K (175-325 °C), more preferably 473-573 K
20 (200-300 °C), and/or at a magnetic field density of 10-50 Gauss.

13. Method according to any of the preceding claims, wherein a frequency of 0.01-10 MHz is used, preferably 0.1-5 MHz, and/or a power of 10-1000 W/cm², preferably 20-500 W/cm²,
25 more preferably 50-250 W/cm², and/or during a period of 1-120 sec, preferably 2-60 sec, and/or at a relative welding speed of 5-200 mm/sec, preferably 10-100 mm/sec.

14. Method according to any of the preceding claims, wherein the panels form a part of transport vehicle, such as
30 an airplane, a car, a truck, a boat, a ship, wherein the part is preferably one of a wing, a tail, a door, a body, a door, a hood, and a frame.

15. Construction element obtainable by a method according to any of the preceding claims, wherein a porosity percentage
35 of the element at the edge zone is $< 7\%$ as determined by ultrasound or computed radiography.

16. Product obtainable by a method according to any of claims 1-12, wherein the product is selected from a part of transport vehicle or the vehicle, such as an airplane, a car,

a truck, a boat, a ship, wherein the part is preferably one of a wing, a tail, a door, a body, a hood, and a frame.

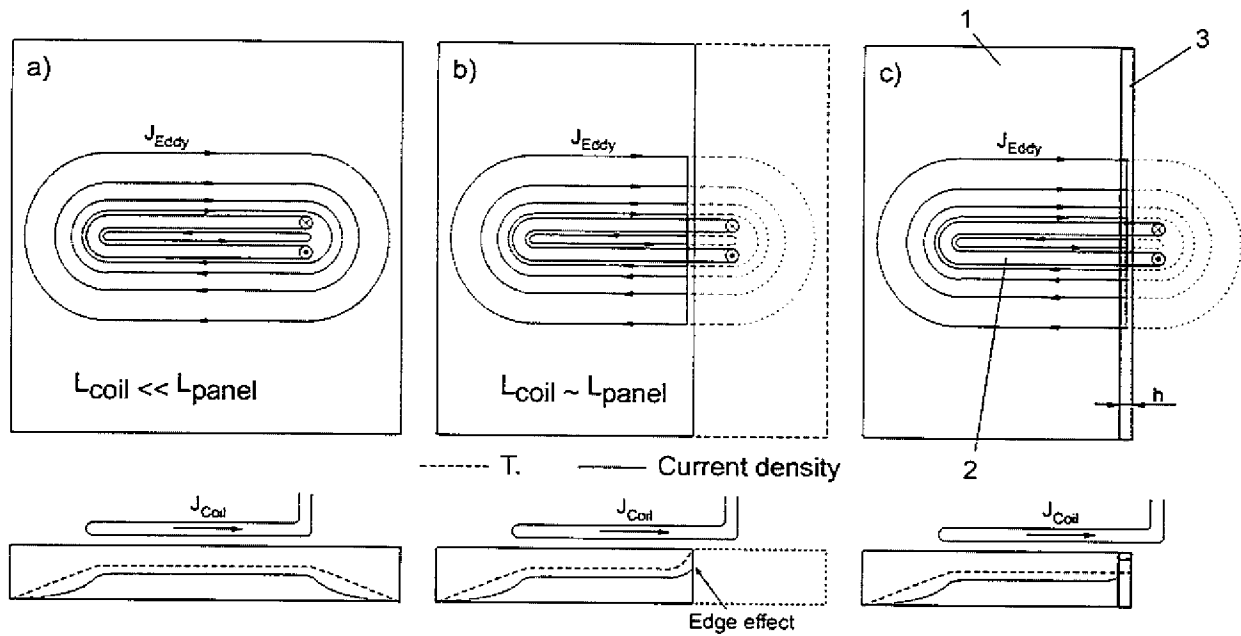
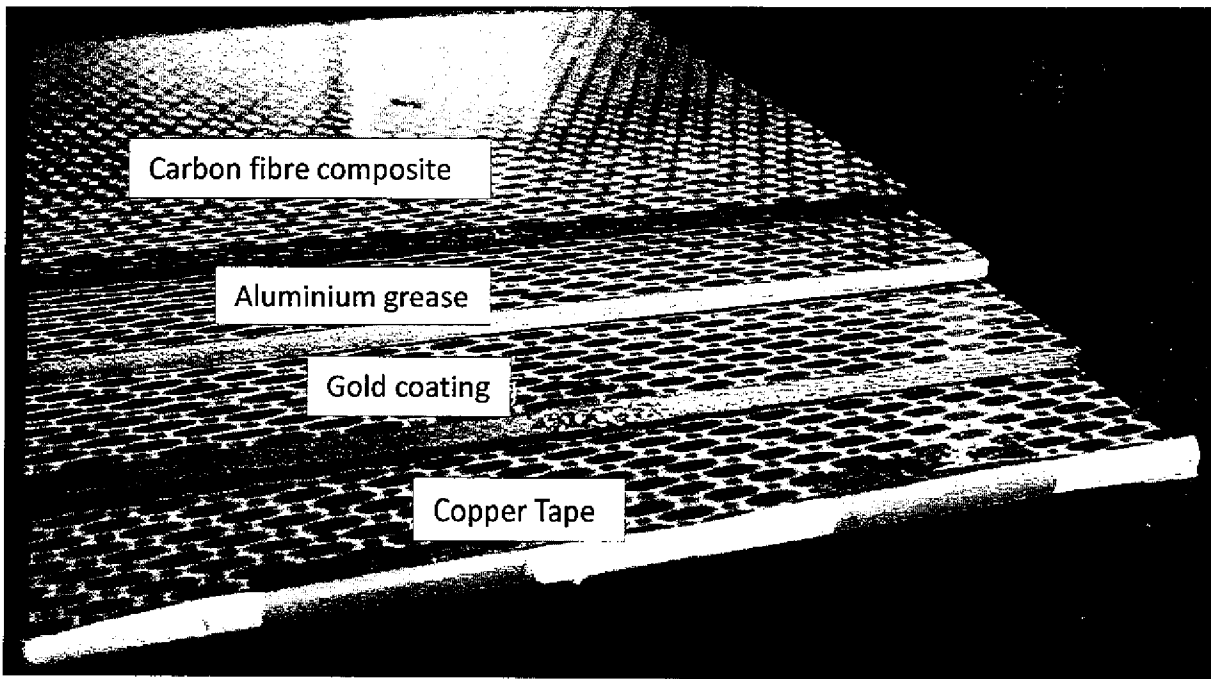


Fig. 2a-c

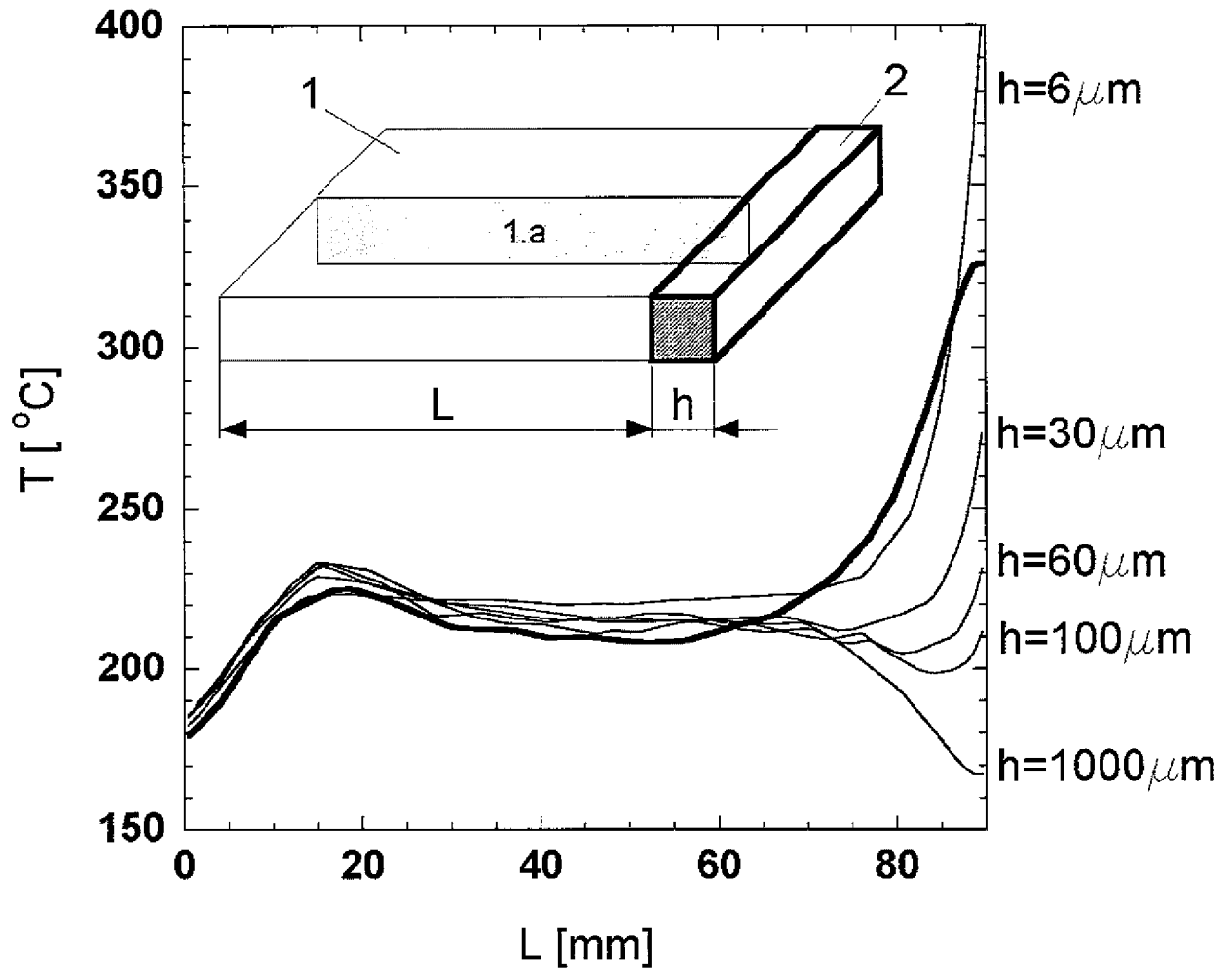


Fig. 3

INTERNATIONAL SEARCH REPORT

International application No
PCT/NL2017/050765

A. CLASSIFICATION OF SUBJECT MATTER
 INV. B29C65/36
 ADD. B29L31/30 B29C65/82

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED
 Minimum documentation searched (classification system followed by classification symbols)
 B29C B29L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
 EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 2013/149501 A1 (PACCHIONE MARCO [DE] ET AL) 13 June 2013 (2013-06-13) paragraph [0003] - paragraph [0077]; figures 2-6	1-16
A	WO 90/08027 A1 (COURTAULDS PLC [GB]) 26 July 1990 (1990-07-26) page 3, line 6 - page 10, line 13; figures 1-5	1-16

Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier application or patent but published on or after the international filing date
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Information on patent family members

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