A COMPREHENSIVE STRATEGY TO INCREASE THE OUT-OF-PLANE THERMAL CONDUCTIVITY OF CARBON FIBER COMPOSITES PROCESSED BY VACUUM ASSISTED RESIN TRANSFER MOLDING

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Abstract

The thermal conductivity of epoxy resin can be increased up to about 20 % by loading it with highly conductive particles, which results in an out-of-plane thermal conductivity of about 20 %, too. Higher loadings increase the viscosity of the resin and hamper its use for liquid composite processes. To derive higher increases in out-of-plane thermal conductivity, additional measures have to be taken. They consist of introducing thermally conductive fibers in out-of-plane direction of the preform by a 3D-weaving process. Measured out-of-plane thermal conductivities of 3D-woven fabric composites show a significant increase compared to a typical laminated composite. An existing analytical model was altered to predict the effective thermal conductivity as a function of the composite material properties such as the thermal conductivities and volume contents of fibers in in-plane and out-of-plane direction, the thermal conductivity of the loaded resin, the grid-density of the out-of-plane fibers, and material properties of the contacting material.

1. Introduction

The ability to remove heat through the thickness of a composite has many possible applications. Leading edges in supersonic aircraft wings, inlet or exhaust areas of gas turbine engines, light weight heat exchangers, electronics packaging materials, hydraulic pump enclosures, and electro-magnetic interference (EMI) enclosures all are subject to localized thermal loads which would preferably be spread out over adjacent cooler areas for subsequent radiation/convection.

Fiber based composites offer the unique ability to tailor material properties locally. Localized matrix or fiber architecture changes can meet local mechanical, electrical, or thermal loads within a component or vehicle, without weight penalty or resort to additional external structures. Such integral materials reduce the number of components leading to not only more elegant designs, but also to structures less costly to manufacture. Local strength, stiffness, toughness, and thermal properties can be tailored using carbon fibers and/or improved matrices. Numerous attempts have been made to increase the thermal conductivity of the matrix [1 – 5]. However, either the loading of the matrix was that high that the viscosity of the resin would prohibit its use in a VARTM process or the thermal conductivity has been raised only up to a factor of about ten. To further enhance the out-of-plane thermal conduvtivity of a composite plate or part, the fiber architecture has to be altered by

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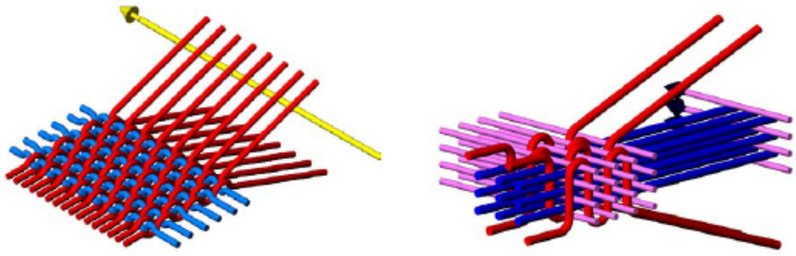
introducing highly conductive carbon fibers in the out-of-plane direction by a 3D-weaving process (Fig. 1) [6].



2-D weave formation 3-D weave formation

Figure 1. Schematic of the 2D and 3D-weaving process

2. Design of Experimental Material

2.1 Objective

It is aimed to produce a composite plate consisting of a 3D-carbon weave and an aluminum-particle filled resin with an elevated out-of-plane thermal conduvtivity. Aluminum was chosen as a good compromise between price and performance.

2.2 Fiber Architecture

The out-of-plane thermal conductivity for laminated composites consists of contributions of vertical z-fibers and the transverse thermal conductivity without vertical fibers. It can be approximated as a parallel connection of both components [7].

$$k_{33} = k_{11}\delta_{z-fibers} + k_{22}(1-\delta_{z-fibers}) \tag{1}$$

with $\delta_{z\text{-fibers}}$ = ν_{Fz}/ν_F
ν_F - fiber volume fraction
ν_{Fz} - fiber volume fraction of z-fibers

The longitudinal in-plane thermal conductivity was described by Thornburgh and Pears [7].

$$k_{11} = k_{Fa}\nu_F + k_M(1-\nu_F) \tag{2}$$

with k_{Fa} - axial fiber thermal conductivity
k_M - thermal conductivity of matrix

The transverse thermal conductivity was modelled by several researchers. A well accepted model was published by Hatta and Taya [8].

$$k_{22} = k_M + \frac{k_M(k_{Fr}-k_M)\nu_F}{k_M+0.5(1-\nu_F)(k_{Fr}-k_M)} \tag{3}$$

with k_{22} - thermal conductivity of cross-ply laminates in transverse direction
k_{Fr} - radial thermal conductivity of fiber
ν_F - fiber volume fraction

Due to the inhomogeneous heat flux through a composite material with high conductive paths in a low conductive material, the influence of sample thickness, grid density of z-fibers and the thermal conductivity of the neighbouring materials have to be taken into account as published in [9], leading to the following equation:

$$k_{33} = k_{11}\xi_{Kmat}\xi_{TD}\delta_{z-fibers} + k_{22}(1-\delta_{z-fibers}) \tag{4}$$

with ξ_{Kmat} - material factor
ξ_{TD} - thickness-grid density factor

2.3 Resin

The thermal conductivity for a particle filled resin can be calculated for particle volume fraction up to 30% with a sufficient accuracy with a model suggested by Maxwell [10].

$$k_{filledmatrix} = k_m \frac{k_p + 2k_m + 2v_p(k_p - k_m)}{k_p + 2k_m - v_p(k_p - k_m)} \tag{5}$$

with v_F - particle volume fraction
k_p - thermal conductivity of particle

For a thermal conductivity of the particle much higher than for the matrix ($k_{aluminum}$ = 166 W/(m K), k_m = 0.2 W/(m K), eqn. 5 can be simplified to

$$k_{filledmatrix} \approx k_m \frac{1 + 2v_p}{1 - v_p} \tag{6}$$

The maximal possible particle volume fraction can be estimated by taking into account the viscosity constraints. For vacuum assisted resin transfer processes, the resin viscosity should not exceed 1 Pas. The increase in viscosity can be described by a model of Krieger and Dougherty [11]:

$$\eta_{filledmatrix} = \eta_m (1 + 2.5v_p + 10.05v_p^2 + 0.00273 \bullet 10^{16.6 v_p}) lg\left(\frac{d_{ref}}{d_p}\right) \tag{7}$$

with η_m - viscosity of neat resin (= 0.2 PA s)
d_p - particle diameter (Al-particle = 4 µm)
d_{ref} - diameter of reference particle (= 100 µm)

For $\eta_{filledmatrix}$ = 1 Pa s computation leads to a maximum particle volume fraction of 15 % and a $k_{filledmatrix}$ of 0.3 W/(m K).

2.4 Material Design

The equations described above can be used to design a composite material with a out-of-plane thermal conductivity of about 10 W/(m K). With the material data given in Chapter 3, the necessary fiber volume fraction of z-fibers can be calculated. With ξ_{Kmat} = 0.60 and ξ_{TD} = 0.48 (aimed sample thickness 5 mm) the necessary fiber volume fraction of the z-fibers can be calculated to be 2.5 % while aiming for an overall fiber volume fraction of 50 %. This means that 95 % of the fibers have to be oriented in in-plane directions and 5 % of the fibers shall be placed vertically to maintain the out-of-plane heat transfer. A 2.5%-content of vertical fibers can be derived by a grid density of about 7 mm.

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3. Experimental

3.1. Materials

3.1.1 Fibers and Resin

For in-plane fibers, the Tenax®-E HTA40 E13 3K 200tex fiber with a thermal conductivity of 17 W/(m K) is used. The out-of-plane fibers were high-modulus pitch-fibers from Mitsubishi (K13D2U 2k 365 tex) with a thermal conductivity of 800 W/(m K).

The matrix material was a cold-curing HP-E3000GL_SDB_resin with HP-E200GL_SDB_hardener from HP-Textiles GmbH in which aluminum particles with an average diameter of 4 µm were distributed. The aimed AL-particle volume fractions were 5%, 10%, 15%, and 20%.

3.1.2 3D-Weaving

A 3D-weaving machine from the company MAGEBA (Fig. 2) was used to weave preforms with thermally high conductive carbon fibers in out-of-plane direction. The in-plane fabric structure was designed similar to a plain weave. The 3D-weaving-machine uses four shuttles to transport weft yarns. The warp-yarns are attached to Jacquard-threads mounted to an Unival from Stäubli, which is able in contrary to conventional Jacquard-devices to lift the warp-yarns to four different height-positions. From 1024 available Jacquard-threads, 256 were used to move the same number of warp-yarns.

Figure 2. 3D-Weaving machine

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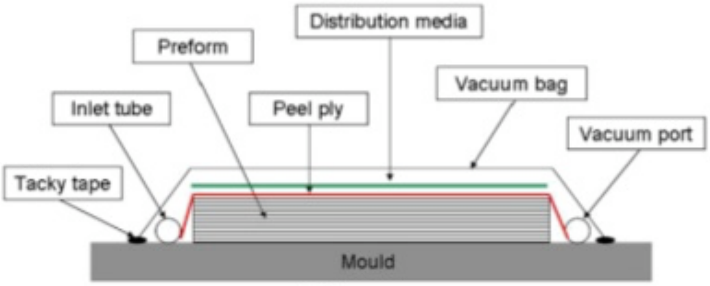
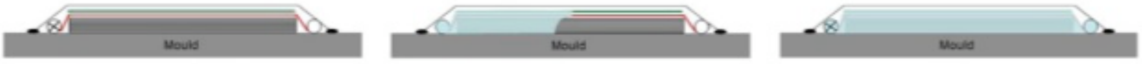
3.2. VARTM

The entire lay-up was assembled in accordance to Fig. 3. Before infusion, the mixed resin was degassed in a pressure pot under vacuum for 10 min to reduce the air content in the resin. The maximum vacuum level was reached after a slow gradual decrease of pressure to avoid boiling of the resin. After degassing, ambient pressure was applied, forcing the resin to move into the still clamped inlet tube. Upon unclamping of the inlet, the resin flowed through the distribution media and impregnated transversely the 3D-carbon preform. Once the flow front had reached the end of preform, the post-filling stage was initiated, pressure at the inlet and vent being set to 500 mbar. Laminate thickness and resin pressure gradients gradually dissipate, and the pressure boundary conditions are maintained until the resin was cured. Subsequently, the cold curing resin was post-cured for seven hours at 65°C.



(a) Lay up



(b) Pre-filling (c) Filling (d) Post-filling

Figure 3. Schematic diagram of stages in the VARTM infusion process [12]

3.3. Thermal Conductivity Measurements

Thermal conductivity measurements were performed with a measuring cell (Figure 4) built in-house at the Institut für Kunststofftechnik Westpfalz according to ASTM E 1225 – 04 allowing measurements of circular samples with a diameter of 50 mm [13, 14].

Sample thicknesses can vary from 2 until 50 mm. Three thermistors in the top meter bar are used to measure heat flux which is assumed to be constant while traveling through the sample into the lower meter bar where a fourth temperature is read. Both meter bars are made out of stainless steel (k_{mat} = 16 W/(m K).The top meter bar is insulated with foam and additionally shielded by a guard heater to prevent radial heat loss. The cell was placed on a water-cooled plate to provide a heat sink with a constant temperature T_{sink} beneath the bottom plate. Thermistor information were evaluated by a LabView-based program. Conductivity paste (OT-201 from Omega with k = 2.3 W/(m K)) was used in order to facilitate coupling and to reduce interfacial thermal resistance. Prior to measurements, the thermal resistance R_{int} at the interfaces from meter bars and sample had to be determined using a stainless steel reference samples. R_{int} was determined to 0.0004150798 (K mm^2)/W. The amount of coupling paste was weighed in order to apply always the same amount. The heat flux introduced by

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the cartridge heater was about 2000 W/m^2. Calibration measurements were performed on isotropic specimens to determine the accuracy of the measuring cell.

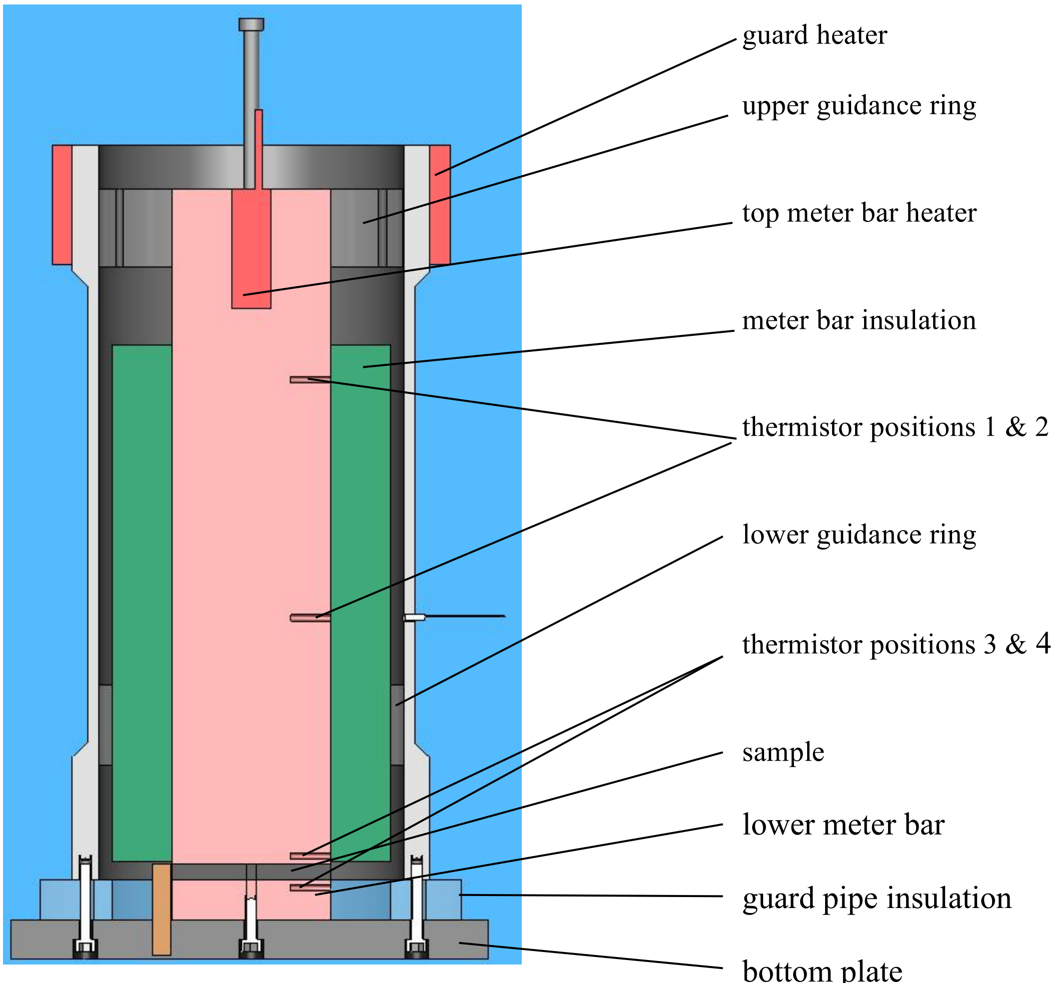

Figure 4. Thermal conductivity measuring device

4. Results

Due to unexpected problems during the VARTM-process, the experimental results can just be presented at the ECCM17 during the presentation.

5. Conclusions

High thermal out-of-plane conductivities in composite parts can be obtained by a combination of improvements in fiber architecture and thermal conductivity of the matrix. However, the dominant role to increase the part´s thermal conductivity is played by the fiber and its architecture. As calculated, using thermally high conductive pitch fibers with small diameters in vertical direction, out-of-plane thermal conductivities above10 W/(m K) can be reached. As shown above, the increase in the thermal conductivity of the matrix is limited. Addionally, the preforms acts as a filter for the dispersed

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particles during the VARTM-process and decrease the particle fraction in the resin and thus its thermal conductivity

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