Pultrusion of Braided Thermoplastic Commingled Yarn – Simulation of the Impregnation Process

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Introduction

Continuous fibre reinforced thermoplastic composites possess many advantages and potentials that are higher than presently utilised in actual applications [1]. Nevertheless, owing to the problems of their impregnation, continuous processing methods for thermoplastic matrices are still difficult to establish for industrial purposes. In our former studies [2], we connected a thermoplastic pultrusion device with a braiding machine in order to realise a continuous manufacturing method for ±45° continuous glass fibre (GF) reinforced pipes with a Polypropylene (PP) matrix. The influence of the parameters preheating temperature and heating die temperature on mechanical and rheological properties was examined. In this paper, two different approaches for modelling of the impregnation process are presented.

Pultrusion combined with braiding

Preimpregnated thermoplastic tapes are too stiff to be braided without damaging the fibres. Thus, flexible prepregs, such as powder impregnated or commingled yarns (polymer fibres mixed with reinforcement fibres) must be used. In this work we applied a GF/PP commingled yarn, named Twintex™ from Vetrotex, Chambery, France. This yarn consists of 35 volume % glass fibres. The diameters of the GF and PP fibres are approximately the same.
The braiding machine was manufactured by Murata Machinery Ltd, Kyoto, Japan, especially for braiding of sensitive rovings. It possesses 32 carriers, 16 of them rotating clockwise and the 16 others counterclockwise around the machine axis.
In Figure 1, the braiding pultrusion line is shown. With the braiding machine two layers of each 32 rovings GF/PP commingled yarn are braided around a PTFE core with a diameter of 10 mm. In the preheating chamber the product is heated to a temperature close to or slightly above the melting point of PP. After the preheating, the product enters the heated die, where the final melting of the polymer and the impregnation of the glass fibres occurs. Then the product is led into the cooled die, which cools the profile under pressure below the melting temperature of the polymer and enables a calibration. The pulling force is applied with a double belt pulling device. After the process, the PTFE core can be pulled out of the profile. Results are pipes with an exterior diameter of 12 mm and a wall thickness of 1 mm.

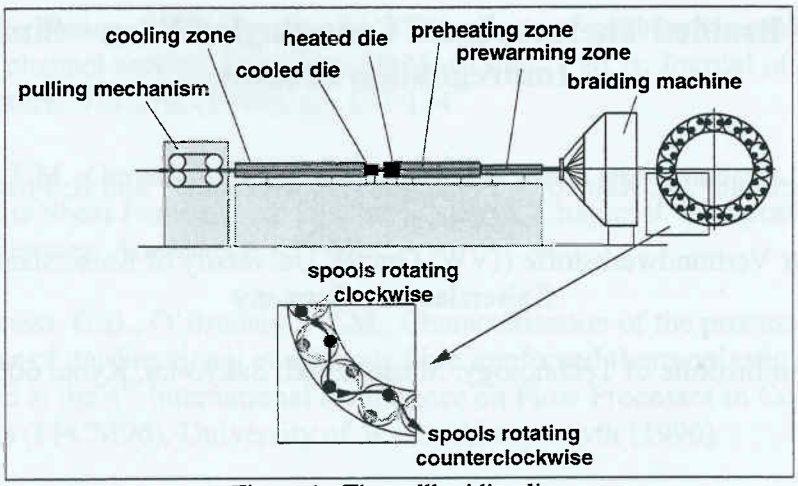

Figure 1: The pullbraiding line

Experimental results

The axial compression modulus of the tube like specimen was found to be the most sensitive property for changes in processing parameters. At a constant pulling speed of 0.1 m/min and a constant heated die temperature of 200°C, the temperature of the preheating zone was varied between ambient temperature and 300°C. As can be seen from Figure 2, a maximum compression modulus is obtained at preheating temperatures around 200°C.

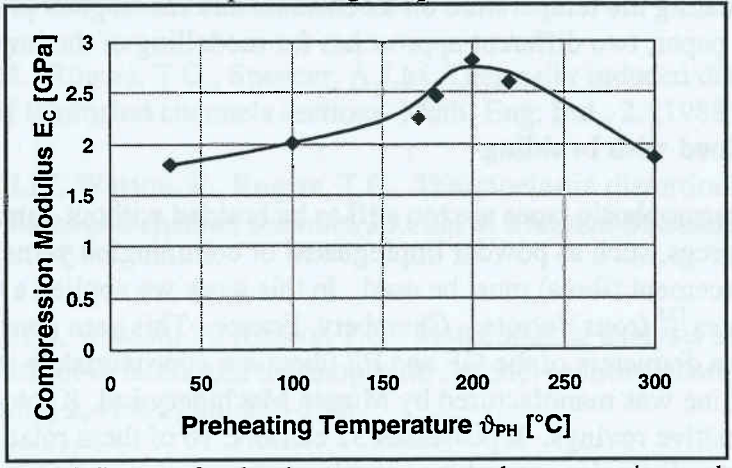

Figure 2: Influence of preheating temperature on the compression modulus

At the same pulling speed and a constant preheating temperature of 220°C, the heating die temperature was varied between the melting temperature of PP, 165°C, and 300°C. Also in this case, a heating die temperature of 200°C turned out to be the optimum condition (Figure 3).

Modelling

Two approaches exist for the physical description of the impregnation of fibre bundles: the micromechanical approach (modelling of the polymer flow around individual fibres) and the continuum mechanical approach (polymer flow through a medium with a continuous flow resistance). For the latter approach, the semi-empirical Darcy's law has widely been used: The vector flow rate $\dot{q}$ is proportional to the gradient of the scalar field of the pressure p.

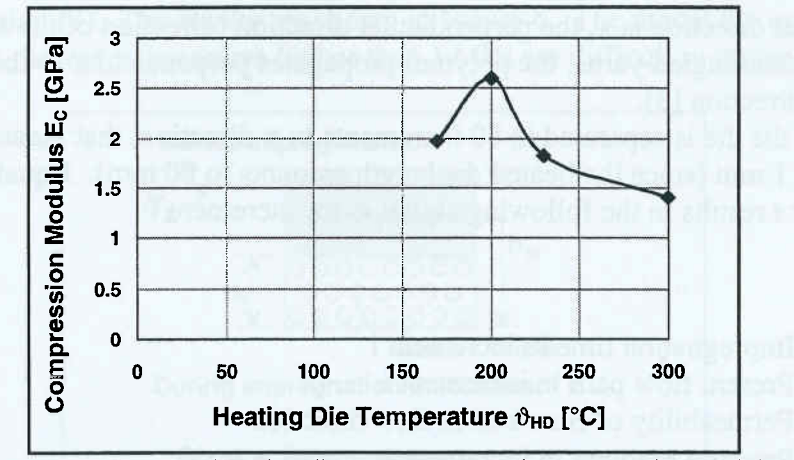

Figure 3: Influence of heating die temperature on the compression modulus

The coefficient of proportionality contains the fluid's viscosity η and the medium's permeability K:

$$\dot{q}_j = -\frac{K_j}{\eta}\frac{dp}{dx_j} = \frac{dx_j}{dt} \tag{1}$$

In spite of the reservations to application of Darcy's law (experimental determination of permeability necessary, law not correct for non-newtonian fluids), no simpler method has gained acceptance yet.

In commingled yarn rovings the polymer fibres and the glass fibres are not distributed homogeneously. The glass fibres are always agglomerated due to the production process. This renders impregnation by the molten polymer difficult. The GF agglomerations have usually an elliptical cross sectional shape with the following typical half axis dimensions:

a - Small GF agglomeration half axis = 33 µm

b - Large GF agglomeration half axis = 59 µm

The GF content inside the GF agglomerations was determined as 0.76, which correspond to a mean number of GF in an agglomeration of 22. The cross section of a quarter agglomeration including proportionate polymer (called 'complete agglomeration' in the following) is then 4472 µm^2, which is equal to a rectangle with the borders 52 µm and 86 µm (Figure 4). At the die exit, this area is decreased to 337.5 µm^2, following the die's geometry. Additionally, the void content should decrease by definition to 2 %.

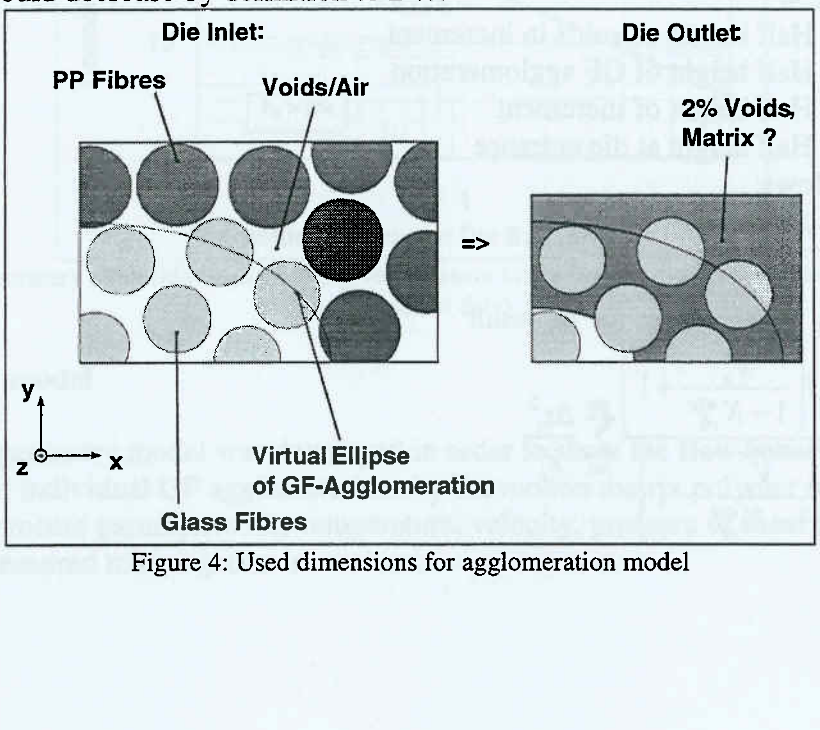

Figure 4: Used dimensions for agglomeration model

The longitudinal direction is x, the perpendicular direction (direction of the small ellipse half axis) is z. In commingled yarns, the polymer propagates perpendicular to the GF axis, therefore in z-direction [3].
For the model, the die is separated in 80 increments in x-direction, that means, every element has a length of 1 mm (since the heated die length amounts to 80 mm). Equation (1) twice integrated after t results in the following equation for increment i:

$$\Delta t_i = \frac{\Delta z_i^2 \eta}{2 K_z \Delta p_i} \tag{2}$$

Δt_i - Impregnation time in increment i
z_i - Present flow path in increment i
K_z - Permeability of fibre bed in flow direction
Δp_i - Pressure gradient in increment i

Gutowski [4] developed an approach to determine the permeability perpendicular to the fibre direction:

$$K_z = \frac{r_f^2 \left(\sqrt{\dfrac{V_a}{1 - X_{v0}^{agg}}} - 1 \right)^3}{4 k_{zz} \left(\dfrac{V_a}{1 - X_{v0}^{agg}} + 1 \right)} \tag{3}$$

r_f - Glass fibre radius
V_a - Maximum possible fibre volume content in GF agglomeration
X_{v0}^{agg} - Void content in agglomeration (related to complete agglomeration)
k_{zz} - Gutowski permeability

The pressure profile can be assumed to be linear [5]. The void content decreases linear with increasing x. The total flow path for each increment is determined geometrically by the rectangular model, shown in Figure 5. Three equations can be determined:

$$h_{vi} = X_{v0}(h_0 - z_i) \tag{4}$$

$$X_{vi} = \frac{h_{vi}}{h_{1i}} \tag{5}$$

$$h_{1i} = h_{10} - X_{v0}^{agg} z_i \tag{6}$$

h_{vi} - Half height of voids in increment
h_0 - Half height of GF agglomeration
h_{1i} - Half height of increment
h_{10} - Half height at die entrance

Thus for z_i follows:

$$z_i = \frac{h_0 X_{v0}^{agg} - h_{10} X_{vi}}{X_{v0}^{agg}(1 - X_{vi})} \tag{7}$$

After inserting the equations for Δt_i result

$$\Delta t = \frac{1}{\Delta p} \frac{2 \eta k_{zz} l \left(\dfrac{V_a}{1 - X_{v0}^{agg}} + 1 \right)}{r_f^2 \left(\sqrt{\dfrac{V_a}{1 - X_{v0}^{agg}}} - 1 \right)^3} \sum_{i=1}^{80} \frac{\Delta z_i^2}{x_i} \tag{8}$$

with z_i from equation (7). The results are shown in Figure 6. The model fits quite well to the experimental data, however, pressures higher than 1 MPa are difficult to obtain in experiments.

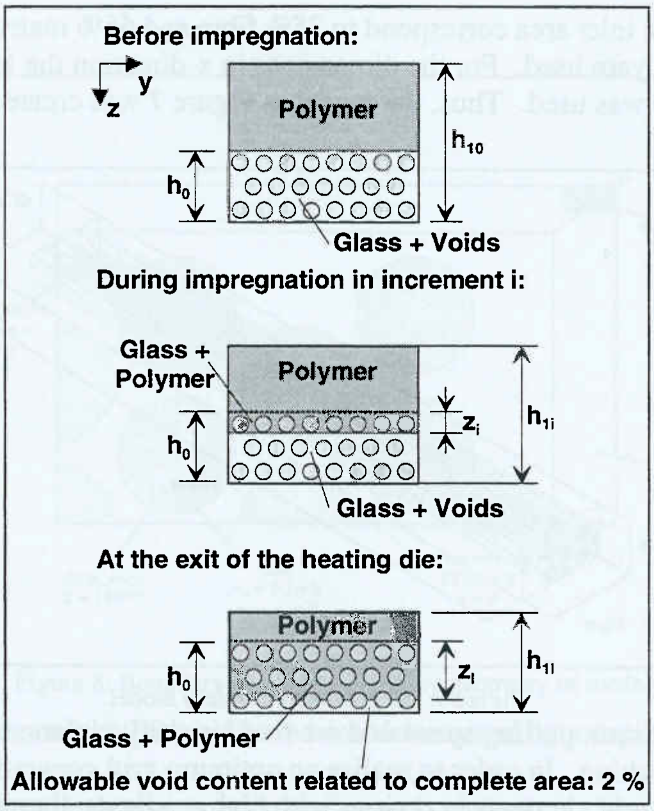

Figure 5: Rectangular model for determination of polymer flow path

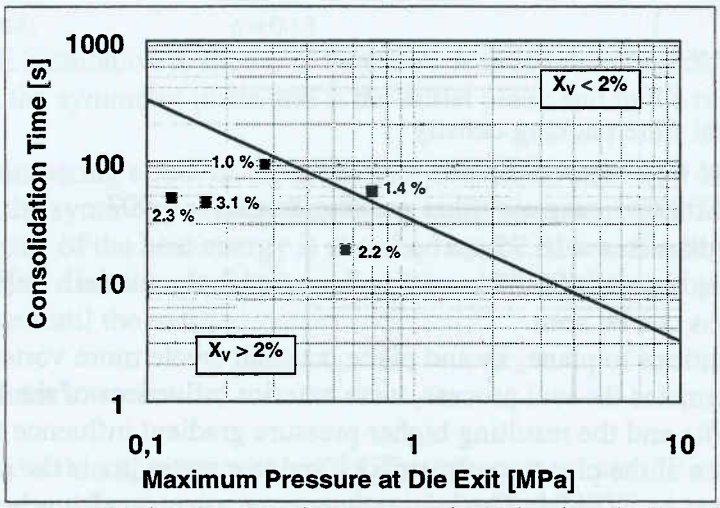

Figure 6: Necessary consolidation time at a given pressure in the heating die (line = model; symbols = experimental data)

Finite element model

The 3D micro-geometry model was developed in order to show the flow behaviour and the impregnation of individual GF agglomerations by the molten matrix polymer in the heated die. Relevant process parameters like temperature, velocity, pressure or shear velocity can be compared to measured macro-geometric values.

At the starting point for the development of the model, similar conditions are used for the analytical model before. Here too, only a quarter section of the elliptical GF agglomeration is simulated.
The dimensions of the inlet area correspond to 35% fibre and 65% matrix area in accordance with the commingled yarn used. For the dimensions in x-direction the length-depth ratio of the applied die cavity was used. Thus, the model in Figure 7 was created.

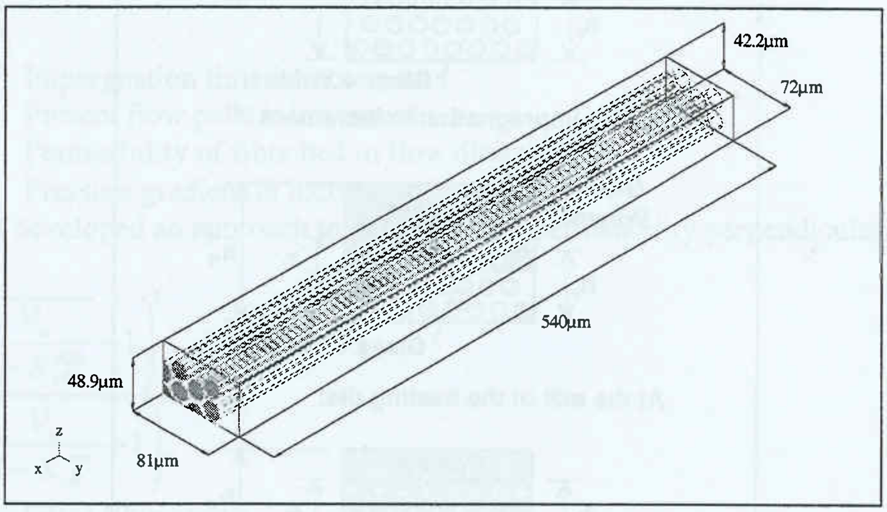

Figure 7: 3d geometry of micro model

The GF possess a constant pulling speed and are fixed in the y-z plane following the hexagonal densest packing. In order to realise an optimum grid generation (numerous small and regular arranged grid elements in regions with high gradients, therefore in the fibre-matrix interface region) a minimum fibre distance had to be kept. Following equation

$$a_F = D_F\left(\sqrt{\frac{\varphi_{max,hex}}{\varphi_{F,local}}} - 1\right) \quad (9)$$

$\varphi_{F,local}$ - Local fibre packing density
a_F - Fibre distance = 3.3 µm
$\varphi_{max,hex}$ - Maximum hexagonal fibre packing density = 0.907
D_F - GF diameter = 16.75 µm

a local fibre volume content of 0.634 was implemented in the model. In Figure 8, the set boundary conditions can be seen.
The adhesion conditions to plane_xy and plane_xz shall create more vortexes in the flow field. This approximates the real process, were exterior influences of the fluid and a bigger taper angle of the die and the resulting higher pressure gradient influence the fluid behaviour. Symmetry condition at the planes symmetry_xy and symmetry_xz is the perpendicular temperature gradient equal zero. The inlet temperature corresponds to the temperature of the preheating zone.
It should be remarked that in the present model, the influence of the ambient air is not taken into account. The matrix is flowing in the model through the whole inlet plane.
The Carreau model [6] was used to describe the viscoelastic-plastic material behaviour of the non-newtonian PP matrix. The dynamic viscosity η dependent on the temperature T and shear velocity $\dot{\gamma}$ can be expressed as follows:

$$\eta = \eta_0\left[1 + (\lambda\dot{\gamma})^2\right]^{\frac{n-1}{2}} \quad (10)$$

with the initial viscosity η_0

$$\eta_0 = a \exp\left(\frac{T_B}{T}\right). \tag{11}$$

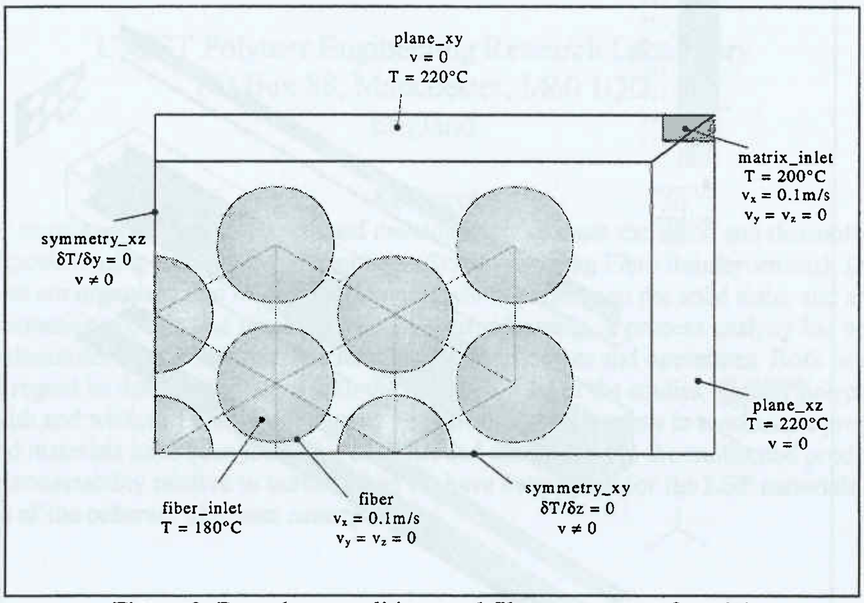

Figure 8: Boundary conditions and fibre geometry of model

The material parameters of the PP used have been determined by a capillary rheometer and a plate rheometer as:

Initial viscosity parameters: $a = 9.12 \cdot 10^{-7}\,\text{Pa} \cdot \text{s}\,; T_B = 9770\text{K}$

Time constant: $\lambda = 0.067\text{s}$

Carreau-index: $n = 0.13$

The result of the FE simulation is shown in Figure 9. It illustrates the temperature distribution of the PP matrix at the symmetry plane and at the outlet plane and in the region of the GF agglomerations.

Noticeable is the hampering effect of the glass fibre bed on the heat flow in the y,z-plane from the outer planes to the symmetry planes. Boundary conditions were 220°C on the outer x,y- and x,z-planes. A part of the heat energy is absorbed by the fibres. During the spreading of the matrix in axial (z-) direction, at first a cooling takes place, followed by a continuous temperature increase until the exit.

Conclusions and future work

The continuous production of pipes with ±45° GF reinforcement and PP matrix is possible. Optimum preheating and heating die temperatures are around 200°C. At the pressures obtained in the heated die, a relatively simple analytical model can be used to determine the necessary consolidation time at a given pressure to obtain an allowable void content of less than 2%. However, to get an impression of the exact temperature distribution, only a Finite Element simulation is meaningful.

The maximum obtained pulling speed is still too low for industrial purposes. Some methods to accelerate the preheating process by a contact heating are tested at present. These experiments will then also lead to higher pressure levels in the heating die, enabling an evaluation of the analytical model for pressures exceeding 1 MPa. Next step of the finite element simulation will be the modelling of the matrix flow containing PP flow fronts driving out the air between the GF.

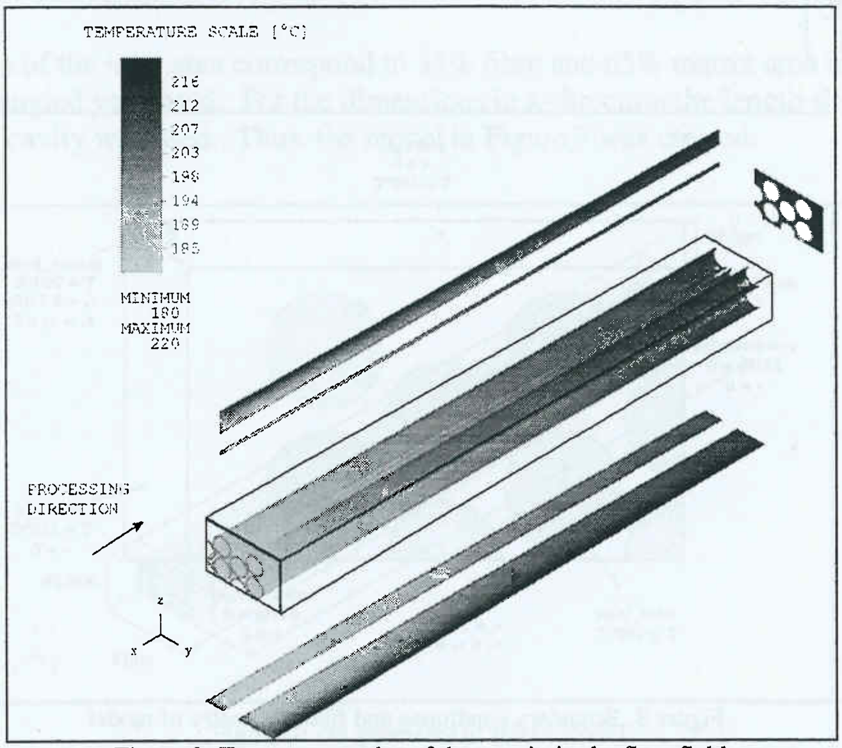

Figure 9: Temperature plot of the matrix in the flow field

Acknowledgements

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