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INFLUENCE OF SPEED OF RESIN INJECTION UNDER PRESSURE INTO MOULD ON STRENGTH PROPERTIES OF POLYMER COMPOSITE

The paper presents a part of research aimed at determining the impact of the speed of resin injection under pressure into the mould on the quality of a laminate. In addition, technological defects such as inaccurate material placement, uneven resin injection into the mould, microvoids and microcracks during gelation are taken into account.For this purpose, the velocity and viscosity of the injected resin (via the Hagen-Poiseuille equation) were analysed analytically, and the gelation temperature of the resin was analysed numerically and compared with the experimental results.Strength tests were carried out on samples of two different lengths made using the RTM method. In the specimens cut with a different measuring base, significant scatter was found resulting from the superposition of effects and phenomena (delamination, voids or scale effect) related to the quality of the produced laminate.

Keywords: strength, laminate, technological parameters, resin medium, RTM method

WPŁYW PRĘDKOŚCI WTŁACZANIA ŻYWICY POD CIŚNIENIEM DO WNĘTRZA FORMY NA WŁAŚCIWOŚCI WYTRZYMAŁOŚCIOWE KOMPOZYTU POLIMEROWEGO

Przedstawiono fragment badań mających na celu określenie wpływu prędkości wtrysku żywicy pod ciśnieniem do formy na jakość laminatu. Dodatkowo uwzględniono wady technologiczne, takie jak niedokładne ułożenie materiału, nierównomierne wtryskiwanie żywicy do formy, mikropustki i mikropęknięcia podczas żelowania. W tym celu przeprowadzono analizę analityczną prędkości i lepkości wtryskiwanej żywicy (poprzez zależność Hagen-Poiseuille'a) oraz analizę numeryczną temperatury żelowania żywicy i porównano ją z wynikami doświadczalnymi. Badania wytrzymałościowe przeprowadzono na próbkach o dwóch różnych długościach, wykonanych metodą RTM. W próbkach wyciętych z różną podstawą pomiarową stwierdzono znaczny rozrzut wynikający z nałożenia się efektów i zjawisk (delaminacji, pustek czy efektu skali) związanych z jakością wytworzonego laminatu.

Słowa kluczowe: wytrzymałość, laminat, parametry technologiczne, medium żywiczne, metoda RTM

INTRODUCTION

In order to obtain a high-quality composite element, it is necessary to control certain parameters such as the injection pressure, outlet pressure, temperature, curing time of the resin medium and mould filling time. This last parameter is particularly important from the industrial point of view. The injection time must be as short as possible, resulting in a completely filled mould [1].

In the initial phase of the moulding cavity filling process (Fig.1), the thermoplastic liquid matrix, at the temperature set for the plasticising system, flows through the central channel, the feed channel, and up to the moulding cavity [1-6].This flow rate depends on the shape and dimensions of the channels and the venturi, as well as on the injection conditions (mainly on the injection speed, pressure and the temperature of the resin medium) and on the properties of the resin mixture, mainly its viscosity [2, 7, 8]. Even a small increase in viscosity of the order of 0.01 kg/(m·s) results in a decrease in velocity of approximately 3 m/s. Considering this problem from the point of view of Darcy's law, the area near the edge will be a channel with a much larger cross-sectional diameter compared to the capillary channels present in the rest of the preform and with virtually unlimited permeability ($k_x \approx 1$) [3].

Attempts to numerically characterise the heat transfer coefficient at the pore size level have shown low accuracy. In general, the preform (or preform layer system) is modelled at the pore size level, which are modelled as cylinders or square rods. The velocity profile is determined and then the local heat transfer between the fabric and fibres is calculated using a non-local thermal equilibrium model. Kuwahara and Nakayama [4] created the model as a closed cell to represent the flow around a cylindrical rod.

Then the volumetric flow of the resin medium is the flow through the circular tubes from the injection port to the outlet port. Additionally, the flow velocity ($\dot{V}$) of this resin medium at selected points of the tube A, B, C and D can be estimated by the Hagen-Poiseuille equation (Fig. 1)

$$\dot{V} = (\frac{\pi r_t^4}{8\mu l})(P_i - P_P - \rho g z) \quad (1)$$

After transforming Formula (1), we obtain (2), determining the viscosity at selected points (Fig. 1)

$$\mu = ((P_i - P_P) - \rho g z)\left(\frac{\pi r_t^4}{8 l \dot{V}}\right) \quad (2)$$

After transforming Formula (1) we obtain (3), defining the change in the duct volume at selected points (Fig. 1).

$$r_t = \sqrt[4]{\frac{\dot{V} 8 \mu l}{(P_i - P_P - \rho g z)\,\pi}} \quad (3)$$

where: $\dot{V}$ – volume flow [m^3/s], r_t – tube radius [m], μ – resin viscosity [kg/(m·s)], l – tube length [m], g – gravity [m/s^2], P_i – absolute pressure in the injection vessel [Pa], P_p – absolute pressure at the inlet port [Pa], ρ – resin density [kg/m^3], z – height difference between the inlet port and fluid level in the injection vessel [m].

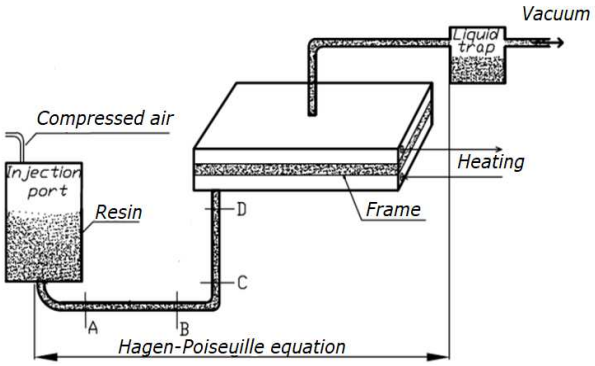

Fig. 1. Diagram of RTM apparatus with marked points at which speed was determined on basis of Hagen – Poiseuille equation

Rys. 1. Schemat aparatury RTM z zaznaczonymi punktami, w których wyznaczona została prędkość na podstawie równania Hagen-Poiseuille

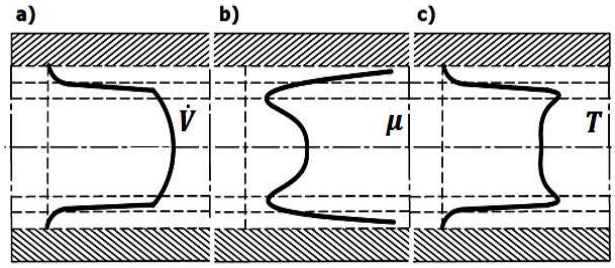

Fig. 2. Characteristic distribution profiles: a) speed, b) viscosity, c) temperature, in supply channel

Rys. 2. Charakterystyczne profile rozkładu: a) prędkości, b) lepkości, c) temperatury w kanale doprowadzającym

The flow velocity ($\dot{V}$) of the liquid resin medium in the set layer is equal to zero at the channel wall and increases slightly as the distance from its inner surface increases (Fig. 2a). In the area where the resin changes to the liquid state, this velocity increases significantly, reaching the highest value in the centre of the channel [9].

Uneven flow conditions of the plastic in the cross-section of the channel are the reasons for changes in its viscosity (μ) (Fig. 2b). At the channel wall, in the area of the most intensive heat exchange between the hot plastic and the channel wall with a lower temperature, the viscosity has the highest value, which for most thermoplastics is about 10^7 Pa·s [10].

The temperature (T) distribution in the channel is influenced in particular by two opposing phenomena, namely heat transfer from the hot plastic to the channel wall and heat generation during flow as a result of internal friction in the plastic and external friction of the liquid plastic against the surface of the solidified layer (Fig. 2c). Near the duct wall, heat generation is exchanged by conduction from the plastic towards the duct wall with a lower temperature [9].

SELECTION OF COMPOSITE COMPONENTS AND ANALYSIS OF PROCESS OF INTRODUCING RESIN MEDIUM IN LIQUID STATE

In order to perform the task, polymer matrix components in the form of 450 g/cm^2 glass matrix and Firestop@ 8170-W1 polyester resin from Bueffa with non-flammable properties were used for the study. The composite was moulded using the RTM (resin transfer moulding) method by injecting the resin mixture between the mould and the dry reinforcement package (preforms). The resin with hardener is injected through a spigot located in the centre of the mould. The two halves of the mould are pressed together with a high closing force.

The resin is injected into the mould under pressure up to several MPa. The reinforcement content in the symmetric composite with the $[0/90/0/90]_S$ arrangement (Fig. 3) was estimated at the level of 60%. The technological parameters of the moulded laminate are presented in Table 1.

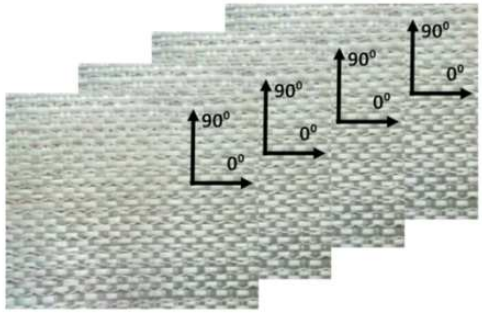

Fig. 3. Way of laying reinforcement in laminate

Rys. 3. Sposób ułożenia wzmocnienia w laminacie

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TABLE 1. **Parameters of composite [0/90]$_s$ formed by RTM method**
TABELA 1. **Parametry kompozytu [0/90]$_s$ formowanego metodą RTM**

Hardener [%]	Demoulding time [h]
Butanox M50,4 with NCL-10.2 initiator	24
Gelation time [h]	Additional heating [°C]
(T = 22-23°C)	30 (16 h)

The 3D fabric-based laminate exhibits the presence of rather large gaps between the fibre strands – resembling a truss-like structure (Fig. 3). Such hollow areas are easily filled with resin with hardener during saturation and do not resist flow [11]. This results in significantly faster saturation than in the case of both classic and stitched preforms.

COURSE OF PROCESS OF INTRODUCING LIQUID RESIN MEDIUM INTO SUPPLY CHANNEL

The resin mixture first flows along the edge of the preform, causing it to "close" in the middle part of the preform. During this process, so-called air traps are formed inside the reinforcement. The flow of the resinous medium changes the technological parameters of the process. Lower values of the flow resistance of the liquid resin with hardener through the "channels" at the edges of the preform change the pressure in the mould and disrupt the saturation process.

That is why it is so important to measure the time dependence of temperature in advance, during the gelation process of the resin mixture (Fig. 4b). After the resin has been fully injected into the mould, the temperature measurement informs us not only about the value of the temperature peak, but also about the range of uniform working of the mixture.

The experimental curve blue line (Fig. 4b) shows three time intervals:

- I time interval (0-17 min) the temperature rise is approx.1.25°C.
- II time interval (17-22 min) the temperature rise is approx.8°C.
- III time interval (22-24 min) the temperature rise is approx.160°C.

A curve based on numerical simulations (Fig. 4a) in the ABAQUS programme red line (Fig. 4b) is presented above the experimental curve. In the numerical process, the described phenomenon occurs much faster and at higher temperatures. Such differences are caused by the fact that the numerical method does not take into account changes in the properties of the injected resin medium.

The mesh used in the calculations, superimposed on the solid element, does not take into account technological processes such as inaccuracy of material placement, material defects, non-uniform flow of resin with hardener during its injection into the mould, the formation of microvoids and microcracks during gelation.

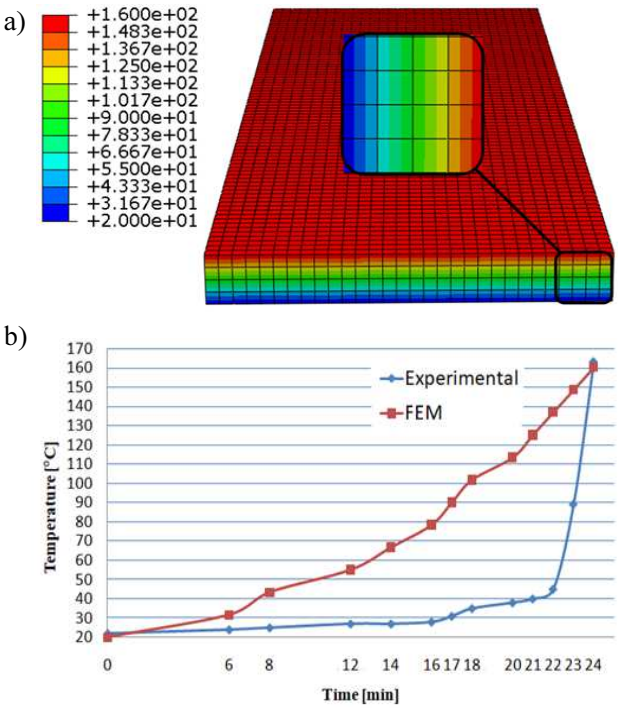

Fig. 4. Numerical simulation of composite gelation (a); graph of composite gelation temperature versus time (b)
Rys. 4. Numeryczna symulacja żelowania kompozytu (a), wykres zależności temperatury żelowania kompozytu od czasu (b)

The distribution of viscosity, flow velocity, shear stress, shear rate and temperature of the liquid medium both in the cross-section and along the channel depends on many factors, primarily on the temperature difference between the channel wall and the resin on the heat transfer coefficient, as well as on the specific heat, density and pressure [8, 12-14].

From the graph presented in Figure 5, it is possible to read the change in velocity at points A, B, C, D and the change in velocity determined on the basis of the Hagen-Poiseuille equation; the range of this relation is presented in Figure 1. Points A, B, C and D represent the decrease in velocity caused by the length of the pipe through which the polymer resin flows (treated as a non-Newtonian fluid). The points presented in Figure 6 show significant decreases in resin velocity, which are caused by small changes in viscosity of the order of 0.01 kg/(m·s) compared to the viscosity specified by the manufacturer [15]. The influence of pressure on polymer viscosity is small. The sensitivity of plastic viscosity to pressure change is expressed by arelative change in viscosity per 1 Pa [12]. For most polymer resins, the ratio of viscosity sensitivity to pressure change and to temperature change is approximately constant and equal to: $-5e^{-7}$,°C/Pa [16].

Pressure is an indispensable parameter in the RTM process. It influences the viscosity of the polymer resin unfavourably as it increases its viscosity (this phenomenon is mainly caused by the mobility of macromolecules, which increases the free volume of the resin), thus decreasing the flow velocity through the supply lines (Table 2 and Fig. 1).

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TABLE 2. **Deformation of resin flow velocity at points a, b, c, d based on formulas (1), (2) and (3)**

TABELA 2. **Wyznaczenie prędkości lepkości przepływu żywicy w punktach a, b, c, d zgodnie z zależnościami (1), (2) i (3)**

Point	Length [m]	Volume flow [m^3/s]	Viscosity [kg/(m·s)]	Wire diameter [mm]
A	0.10	15	0.1054	0.2
B	0.15	10	0.1054	0.2
C	0.20	7	0.112	0.1996
D	0.25	6	0.1054	0.2
Dispersion	-	9	0.007	0.0004
Viscosity based on Hagen--Poiseuille equation	1	1.5	0.126	0.2091

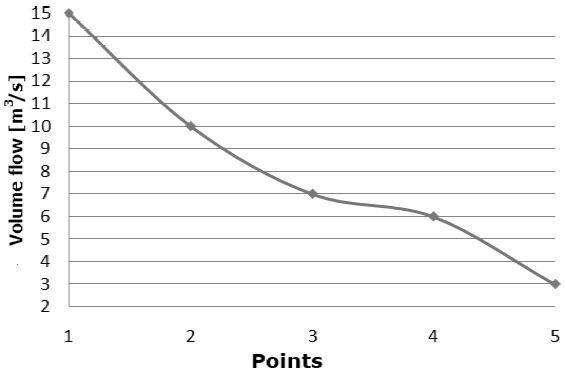

Fig. 5. Resin flow rate in RTM process

Rys. 5. Prędkość przepływu żywicy w procesie RTM

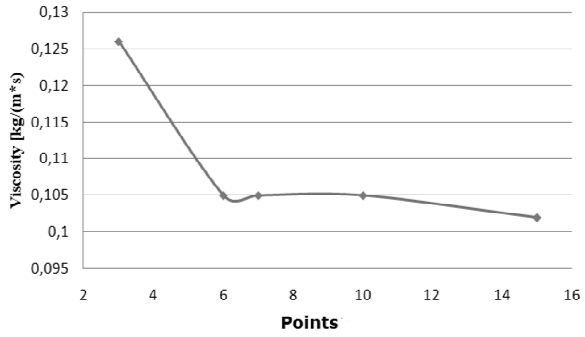

Fig. 6. Resin flow viscosity in RTM process

Rys. 6. Lepkość przepływu żywicy w procesie RTM

The RTM process uses heated moulds (Fig. 1) as increasing the temperature of the resin reduces its viscosity and therefore, faster filling of the dry reinforcement.

Figure 7 shows the change in resin pressure owing to the resistance created by the material placed inside the mould; the graph shows that in the range of 5 to 20 minutes the resin pressure stabilizes. This is because the resin velocity decreases due to the porosity of the perform.

During the cycle of the injection moulding process, heat transfer occurs between the material, the mould, the injection moulding machine and the environment, and heat can be transferred by convection and radiation [15, 17, 18]. In the injection moulding process, heat transfer between the liquid polymer and the surface washed by it is important. During the movement of liquid polymer in its deeper layers, the heat transfer occurs mainly by convection, while in the zone adjacent to the wall – by conduction. Heat transfer by radiation plays a smaller role here [15, 19].

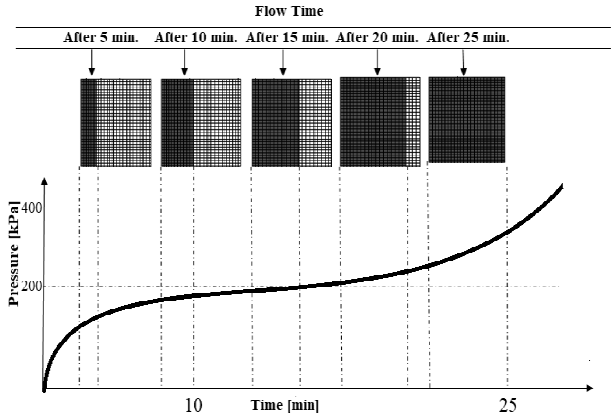

Fig. 7. Resin flow in composite material produced by RTM method

Rys. 7. Przepływ żywicy w materiale kompozytowym wytwarzanym metodą RTM

In the injection moulding process, unsteady heat conduction occurs during both the heating of the polymer in the plasticising system and its cyclic heating and cooling in the mould. When analysing the thermal phenomena taking place in the injection mould, it is necessary to separate the processes taking place in the central runner, feed runner, venturi and moulding cavity. In the analysis of the polymer flow in the moulding cavity, it is usually assumed that in the plane of the moulded part parallel to the polymer flow direction, the heat is transferred by convection, while in the direction of its thickness – by conduction [20, 21].

In the filling phase, due to the high flow velocity of the polymer, the share of convection heat is much higher than that resulting from conduction. In contrast, conduction plays a dominant role in the pressure phase, where the flow velocity is small or even negligible [14].

The cured polyester resin contains not only unbound saturated polyester molecules, but also unbound unsaturated polyester molecules that have not entered into copolymerisation reactions. Thus, the cured resin contains a greater or lesser amount of unbound polyester, which can be assumed to cause some deterioration in its mechanical properties as well as chemical and heat resistance [22].

The heterogeneity of the resin injection process has a significant effect on the strength scatter (Table 3).

Specimens were cut out of the fabricated laminate by CNC milling, according to ISO 527-4:1997. The layout of the specimens is shown in Figure 8. The process is described in detail in article [23].

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TABLE 3. **Values of constant parameters**
TABELA 3. **Wartości parametrów stałych**

Parameter	Value
Press-in pressure [kPa]	295
Pressure in mould [kPa]	4.482
Resin viscosity [kg/(m·s)]	0,09
Time for resin to flow through mould [s]	1500
Time of resin flow into mould [s]	5

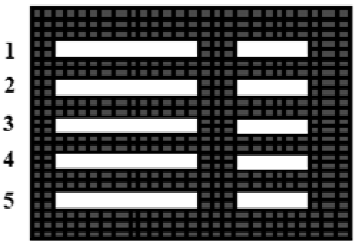

Fig. 8. Diagram of plate fragment with specimens cut out (numbering
Rys. 8. Schemat fragmentu płyty wraz z wyciętymi próbkami (numeracja dotyczy zarówno próbek dużych, jak i małych)

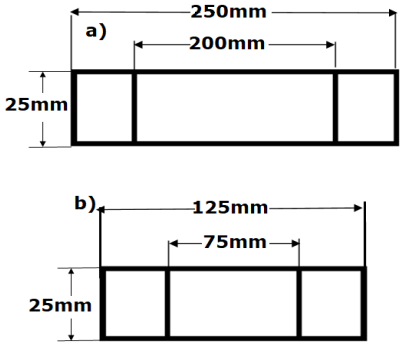

Fig. 9. Dimensions of specimens: a) long specimen, b) short specimen applies to both and long specimens
Rys. 9. Wymiary próbek: a) próbka długa, b) próbka krótka

Subsequently, the specimens(smaller and larger with the dimensions shown in Fig. 9) were subjected to static tensile testing on an Instron 8501 testing machine to determine the measure of composite failure [24]. Defects such as delamination, microvoids and scale effects that are directly related to the quality of the produced laminate were taken into account. The data obtained from the experiment are summarised in Table 4.

The scientific literature contains very little information on the influence of the scale effect on the elastic and plastic properties of a material. This is due to the complexity of the experiments and the great difficulty in interpreting the results. In a composite, it being a linear elastic or viscoelastic material, complicated processes of stress distribution take place, the values of which depend on the heterogeneous structure of the composite and on the type of defects occurring in it (microvoids and microcracks) [24].

The likelihood of more defects appearing in a longer sample than in a shorter sample as a result of scale effects is due to the changing viscosity values of the resin mixture. The main cause of this phenomenon is the mobility of the macroparticles, which increases the free volume of the resin medium, thus lowering the velocity of the matrix through the feed lines.

TABLE 4. **List of values for specimens subjected to tensile test**
TABELA 4. **Zestawienie wartości dla próbek poddanych próbie rozciągania**

Specimens made by RTM (long)	σ [MPa]	Specimens made by RTM (short)	σ [MPa]
1	18.21	1	20.40
2	18.32	2	20.21
3	18.13	3	20.33
4	18.10	4	20.13
5	18.40	5	20.20
Average	18.23	Average	20.25
Dispersion	0.325	Dispersion	0.273

CONCLUSIONS

The speed of injection of the resin under pressure into the mould is one of the most important technological parameters in the RTM method.

An uneven flow velocity of the resin with hardener (e.g. at the injection stage through the circular tubes from the injection port to the port connecting the cavity with the mould) causes significant decreases in the flow velocity of the resin mixture owing to an increase in resin viscosity and gelation temperature.

The reason for the above phenomena is mainly a result of the mobility of macromolecules, which will increase the free volume of the resin medium.

Analyses of the cut laminate plate specimens with different measurement bases prove that the superposition of effects and phenomena (delamination, voids, or scale effect) related to the quality of the manufactured laminate, deteriorates the mechanical properties of the material.

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