

RIGA TECHNICAL UNIVERSITY

Faculty of Civil Engineering
Institute of Materials and Structures

Pāvels Akišins

Doctoral Student of the Study Programme “Civil Engineering”

DESIGN OF ADVANCED PULTRUSION PROCESSES

Summary of the Doctoral Thesis

Scientific supervisor
Professor Dr. sc. ing.
JEVGENIJS BARKANOVŠ

RTU Press
Riga 2017

Akišins, P. *Design of Advanced Pultrusion Processes*. Summary of the Doctoral Thesis. Riga: RTU Press, 2017. 38 p.

Published in accordance with the decision of the Promotion Council “P-03” of 21 September 2017, Minutes No. 2017-3.



This work has been supported by the European Commission under the Seventh Framework Program, Contract No. NMP2-SL-2013-609149, project “Development of an Innovative Manufacturing Process for the In-line Coating of Pultruded Composites (COALINE)”

ISBN 978-9934-22-023-4

DOCTORAL THESIS PROPOSED TO RIGA TECHNICAL UNIVERSITY FOR THE PROMOTION TO THE SCIENTIFIC DEGREE OF DOCTOR OF ENGINEERING SCIENCES

To be granted the scientific degree of Doctor of Engineering Sciences, the present Doctoral Thesis has been submitted for the defence at the open meeting of RTU Promotion Council on December 19, 2017 at Riga Technical University, 1 Kalku Street, Room 119.

OFFICIAL REVIEWERS

Professor Dr. habil. sc. ing. Lothar Kroll
Technische universitat Chemnitz, Germany

Dr. Ferran Marti
AIMPLAS-Instituto Tecnologico Del Plastico, Spain

Professor Dr. habil. sc. ing. Vladimirs Kasjanovs
Riga Stradins University, Latvia

Professor Dr. sc. ing. Igors Tipāns
Riga Technical University

DECLARATION OF ACADEMIC INTEGRITY

I hereby declare that the Doctoral Thesis submitted for the review to Riga Technical University for the promotion to the scientific degree of Doctor of Engineering Sciences is my own. I confirm that this Doctoral Thesis had not been submitted to any other university for the promotion to a scientific degree.

Pāvels Akišins (signature)

Date:

The Doctoral Thesis has been written in English. It consists of Introduction, 7 Chapters, Conclusion, 123 figures, 23 tables, 1 appendix; the total number of pages is 105. The Bibliography contains 78 titles.

CONTENTS

1. REVIEW OF THESIS.....	5
1.1. Topicality of the Theme	5
1.2. Aims of the Thesis.....	6
1.3. Research Objectives of the Thesis.....	6
1.4. Research Tools and Methods.....	6
1.5. Scientific Novelty of the Thesis	6
1.6. Practical Value of the Thesis	7
1.7. Approbation of Thesis Results and Publications.....	7
2. Structure of the Thesis.....	8
2.1. Literature Review	8
2.2. Modelling of Conventional Pultrusion Processes.....	9
2.3. Curing Kinetic Modelling.....	13
2.4. Experimental Validation of Simulation Algorithms.....	16
2.5. Modelling of Microwave Assisted Pultrusion Processes	19
2.6. Design of Multifunctional Pultrusion Die	24
2.7. Optimisation of Microwave Assisted Pultrusion Process for Multifunctional Die ...	27
2.8. Conclusions	33
REFERENCES.....	35
3. Approbation and publications	37
3.1. List of Publications	37
3.2. List of Conferences.....	37
3.3. Project Seminars	38
3.4. Approbation of Thesis Results	38

1. REVIEW OF THESIS

1.1. Topicality of the Theme

Pultrusion is a continuous and cost-effective process for a production of fibre-reinforced polymer composite structural components with a constant cross-sectional profile (Fig. 1.1). It is an automated process requiring little labour and produces minimal waste material. Conventional pultrusion process that uses electric heaters for heating of the pultrusion die is known since 1951, then in was patented [8]. In 2012, the volume of composites used in pultrusion processes sums up to 47 000 t, covering near 20 % of the total volume of currently used thermoset composites produced in Europe [23]. During the last 5 years, the pultrusion market has experienced significant growth especially in transportation sector and construction applications and is expected to reach \$ 1.7 billion in 2017. According to a new market report [9], the future of the global pultrusion market looks bright with the growth at a CAGR of 4.6 % from 2016 to 2021. The major growth drivers for this market are the rise in demand for lightweight materials and increasing demand for durable products for corrosive environments, such as in rebar and grating applications. These drivers make pultrusion one of the fastest growing manufacturing processes within the composites market.

To improve the effectiveness of conventional pultrusion processes (Fig. 1.2) and quality of pultruded coated profiles, development of microwave assisted pultrusion process with in-line coating technology (Figs 1.3 and 1.4) considerably reducing the number of manufacturing steps is proposed.

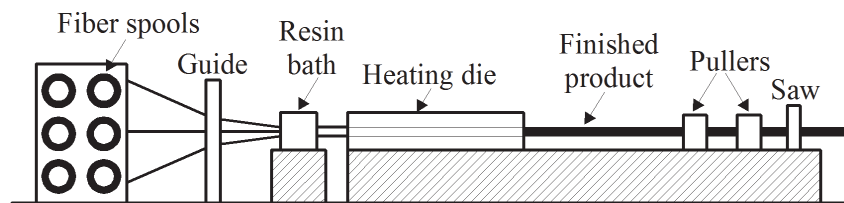


Fig. 1.1. Conventional pultrusion tool.

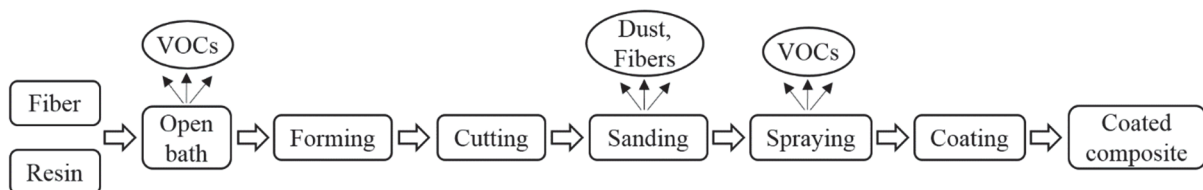


Fig. 1.2. Conventional manufacturing of coated pultruded profile.

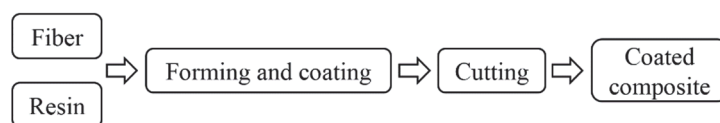


Fig. 1.3. Advanced manufacturing of coated pultruded profile.

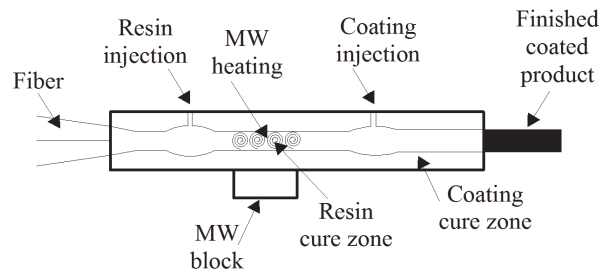


Fig. 1.4. Scheme of advanced multifunctional pultrusion die.

1.2. Aims of the Thesis

The main aim of the present Thesis is the development of new methodology and tools for simulation, design and optimisation of the advanced microwave assisted pultrusion process with in-line coating technology. This methodology is based on the joint use of different finite element codes, planning of experiments and response surface technique and allows to obtain the parameters of technological process with minimal energy consumption.

1.3. Research Objectives of the Thesis

- Development of holistic numerical procedure for a simulation of pultrusion processes.
- Development of curing kinetic models for the resins with high microwave absorption properties.
- Experimental validation of the numerical simulation procedures developed in the Thesis.
- Development of reliable numerical procedure for a simulation of microwave assisted pultrusion processes.
- Design of microwave assisted pultrusion process for a multifunctional pultrusion die.
- Optimisation of microwave assisted pultrusion process with in-line coating technology.

1.4. Research Tools and Methods

Research methods used in the present Thesis include the following:

- finite element software ANSYS Mechanical, ANSYS CFX, COMSOL Multiphysics;
- APDL programming;
- methods of experimental design and response surface technique;
- EDAOpt software for experimental design, research and optimisation;
- solver tool of Microsoft Excel program for a solution of optimisation problems;
- graphical tools of Microsoft Excel program for presentation and analysis of the results.

1.5. Scientific Novelty of the Thesis

In the present Thesis, new simulation and optimal design methodology of advanced microwave assisted pultrusion process with in-line coating technology is proposed. The developed methodology is based on the join use of different finite element codes for solution of coupled thermochemical and electromagnetic problems, and optimisation software for process optimisation. As a result of numerical study, recommendations for designers of

advanced pultrusion processes and multifunctional dies are formulated. Based on results of optimisation, a Microsoft Excel tool is developed for an effective design of the technological process. Using this tool, technologists can find parameters of the pultrusion process for the manufacturing of qualitative coated profile using minimal energy amount.

1.6. Practical Value of the Thesis

The results of this Thesis are a part of the collaborative European project “Development of an Innovative Manufacturing Process for the In-Line Coating of Pultruded Composites” (COALINE) under FR7 program. The main aim of the project is to develop an innovative manufacturing process for the in-line coating of pultruded composites. The data obtained from investigations are intended for the most experienced European companies, research institutes and academic institutions working in the field of pultrusion. The development of new simulation and optimisation methodologies for the design of advanced pultrusion processes will allow further expansion of investigations in this field.

1.7. Approbation of Thesis Results and Publications

The results of the present Thesis have been presented at seven international conferences and eight working meetings within the COALINE project. The Doctoral Thesis has been presented and discussed at scientific meeting in the Institute of Mechanics, Riga Technical University. Main results of the Thesis have been reflected in six papers published in international journals and conference proceedings.

2. STRUCTURE OF THE THESIS

The present Thesis consists of eight chapters, which are organized as follows:

Chapter 1 presents general overview on the pultrusion processes, their simulation and optimisation;

Chapter 2 presents general formulation and numerical implementation of the thermochemical analysis of pultrusion processes. Two numerical procedures based on different finite element codes (ANSYS Mechanical and ANSYS CFX) are developed for a solution of thermochemical problem. The validation of these procedures by experimental and numerical results published in scientific literature is also presented;

Chapter 3 presents the development and validation of methodology for building of curing kinetic models for thermoset resins with high microwave absorption properties. The developed methodology has been implemented in a Microsoft Excel tool;

Chapter 4 presents experimental validation of the developed procedures for numerical simulation of pultrusion processes. The influence of different process parameters and die material properties on temperature and degree of cure in composite material is shown. Correction of initial data using experimental and simulation results is performed with the aim to obtain reliable results;

Chapter 5 is devoted to the numerical simulation and study of advanced microwave assisted pultrusion process. The influence of capacity of the absorption energy field, application of thermal insulation on the die, preheating of steel die, heating of resin bath and reduction of the steel die length on the parameters of pultrusion process is studied with the purpose to increase its effectiveness;

Chapter 6 is devoted to the design of multifunctional pultrusion die used in the advanced pultrusion process with microwave assisted heating and in-line coating technology;

Chapter 7 describes the optimisation methodology, based on the planning of experiments and response surface technique for optimisation of advanced microwave assisted pultrusion process with in-line coating technology.

Finally, the research achievements of the present Thesis are summarized in **Chapter 8**. Recommendations for the future work are also discussed.

2.1. Literature Review

This chapter presents a comprehensive literature review on the investigation of pultrusion processes. Two main topics are covered in the review. The presents literature overview on simulation of conventional and microwave assisted pultrusion processes. Brief review of methods used in the experimental investigation of pultrusion processes is also given. The review also provides literature review about optimisation of pultrusion processes. The existing literature is classified into three following sections: experimental testing, numerical simulation, and optimisation of manufacturing process.

Conventional pultrusion processes, their simulation and optimisation are widely considered in the scientific literature. Advanced microwave assisted pultrusion processes are studied considerably less, no optimisation problems have been solved for this new technological process. The design of pultrusion dies with in-line coating technology is not mentioned in scientific literature.

2.2. Modelling of Conventional Pultrusion Processes

To numerically study pultrusion processes, the following thermochemical problem consisting of three governing equations should be solved:

$$\left\{ \begin{array}{l} \rho c_p \frac{\partial T}{\partial t} - \frac{\partial}{\partial x} \left(k_x \frac{\partial T}{\partial x} \right) - \frac{\partial}{\partial y} \left(k_y \frac{\partial T}{\partial y} \right) - \frac{\partial}{\partial z} \left(k_z \frac{\partial T}{\partial z} \right) - q_b = 0; \\ \bar{\rho} \bar{c}_p \left(\frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} \right) - \frac{\partial}{\partial x} \left(\bar{k}_x \frac{\partial T}{\partial x} \right) - \frac{\partial}{\partial y} \left(\bar{k}_y \frac{\partial T}{\partial y} \right) - \frac{\partial}{\partial z} \left(\bar{k}_z \frac{\partial T}{\partial z} \right) - q = 0; \\ \left(\frac{\partial \alpha}{\partial t} + u \frac{\partial \alpha}{\partial x} \right) = R_r, \end{array} \right. \quad (2.1)$$

where

- T temperature, °C;
- ρ density of the tooling material, kg/m³;
- c_p specific heat of the tooling material, J/(kg °C);
- k_x, k_y, k_z thermal conductivities of the tooling material in x, y, z directions, J/(m °C s);
- q_b rate of energy exchange at the boundary, J/(m³ s);
- u the pull speed, m/s;
- $\bar{\rho}$ the lumped density for the composite material, kg/m³;
- \bar{c}_p the lumped and specific heat for the composite material, J/(kg °C);
- $\bar{k}_x, \bar{k}_y, \bar{k}_z$ the lumped thermal conductivities of the composite material in x, y, z directions, J/(m °C s);
- q the generative term related to the internal heat generation due to the exothermic resin reaction, J/(m³ s);
- $\alpha = H(t)/H_{tr}$ the degree of cure;
- $H(t)$ the amount of heat evolved during the curing up to time t , J/kg;
- H_{tr} the total heat of reaction, J/kg.

It is necessary to note that the first equation in the system presents the energy equation for the tool, the second – the energy equation for the composite moving in the pull direction and the third – the species equation (transport equation) for the resin.

Heat transfer in the composite occurs as a result of conduction and the generation of heat resulting from the exothermic chemical reaction initiated by the die temperature. The generative term related to the internal heat generation due to the exothermic resin reaction could be written as:

$$q = V_r \rho_r H_{tr} R_r, \quad (2.2)$$

where V_r is the resin volume fraction; ρ_r is the resin density, kg/m³;
 R_r is the rate of resin reaction, 1/s, determined as:

$$R_r(\alpha, T) = \frac{\partial \alpha}{\partial t} = \frac{1}{H_{tr}} \frac{dH(t)}{dt} = K(T) \cdot f(\alpha), \quad (2.3)$$

where $f(\alpha)$ depends on the resin properties and varies with the applied resin reaction model;
and $K(T)$ is defined by the Arrhenius relationship:

$$K(T) = K_0 \exp\left(-\frac{E}{RT}\right), \quad (2.4)$$

where $R = 8.314$ is the universal gas constant, (J/(mol °C)); E is the activation energy, J/mol;
and K_0 is the frequency factor, 1/s.

Two numerical procedures for the simulation of the pultrusion process were developed. The first procedure was developed in ANSYS Mechanical environment and was based on the mixed time integration scheme and nodal control volume method to solve simultaneously the coupled temperature state and degree of cure by using an iteration technique. A uniform finite element discretization of the pultruded composite profile is applied in the pull direction. The nodal control volumes are constructed based on the finite element mesh as presented in Fig. 2.1. The centres of the control volumes coincide with the nodal points of the finite element. In the control volume the distribution of a field variable is assumed constant and its value is defined by the field variable calculated at the representative finite element node.

At the beginning it is assumed that the degree of cure has the same value α^0 in each nodal control volume of the composite. In most cases it equals zero. Then the transient thermal finite element analysis is performed to obtain an initial state of temperatures for each element. From the temperature field, the rate of cure for the nodal control volume j at any type step i is calculated outside the finite element software by the user developed program.

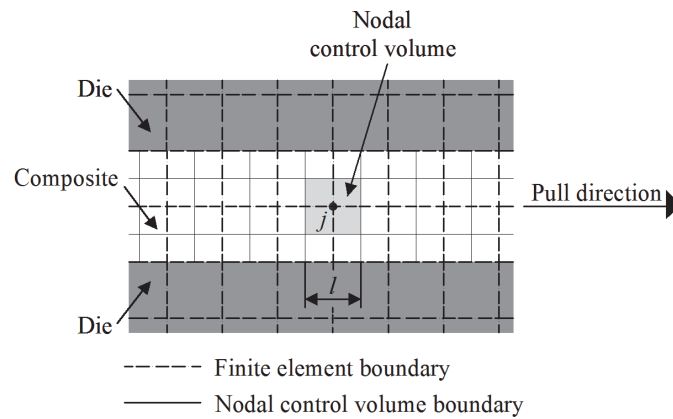


Fig. 2.1. Finite element and nodal control volume meshes.

$$\frac{\partial \alpha_j^i}{\partial t} = \left[\frac{\Delta \alpha_j^i}{\Delta t} \right] = K_0 \exp \left(-\frac{E}{RT_j^i} \right) f(\alpha_j^{i-1}). \quad (2.5)$$

For $t > 0$ s, the degree of cure can be obtained continuously by using the following relation:

$$\alpha_j^i = \alpha_j^{i-1} + \left[\frac{\Delta \alpha_j^i}{\Delta t} \right] \Delta t, \quad (2.6)$$

where Δt is the time step, s, determined as:

$$\Delta t = \frac{1}{p} \cdot \frac{l}{u}, \quad (2.7)$$

where l is the length of nodal control volume in the pull direction, m; u is the pull speed, m/s; and p is the number of sub-steps. If the procedure of sub-stepping is not applied, $p = 1$.

The exothermic effects of cure reaction are evaluated as the equivalent nodal heat power for a nodal control volume or node j by the following relation:

$$q = V_r \rho_r H_r R_r = V_r \rho_r H_r \left[\frac{\Delta \alpha_j^i}{\Delta t} \right]. \quad (2.8)$$

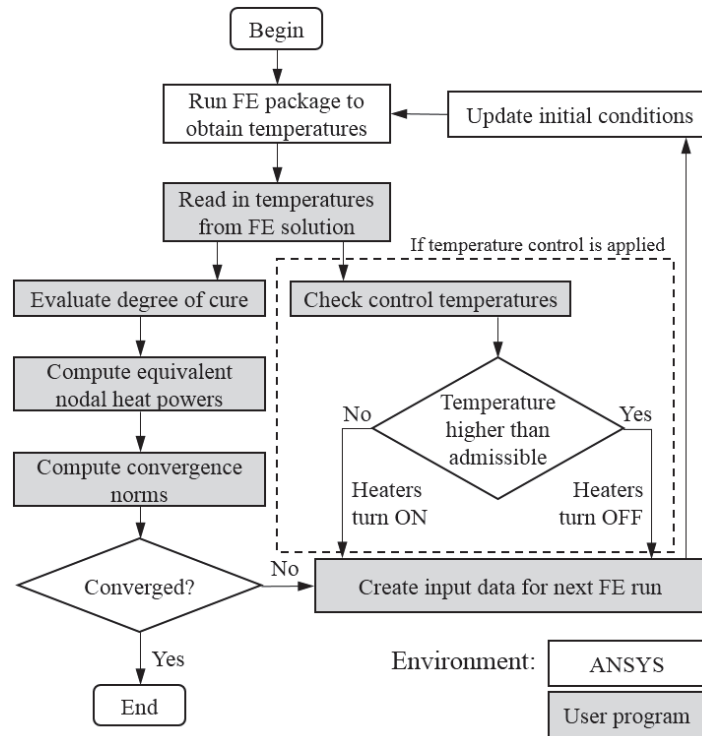


Fig. 2.2. Flowchart of the numerical procedure based on ANSYS Mechanical.

These values will be applied to calculate the temperature field for a new step of iteration. Thereby, a movement of the resin-saturated composite is simulated by shifting the temperature and degree of cure fields after each calculation step. It is necessary to note that at the entrance of the die, the degree of cure remains unchanged and equals to α^0 at any step of iterations. In general, the algorithm can be summarized as a flowchart presented in Fig. 2.2.

The second procedure has been developed by using ANSYS CFX software. The composite is modelled as a heterogeneous mixture of two components with the same physical characteristics which represent the reacted and unreacted material. The degree of cure α is modelled as a mass fraction of the reacted substance. During the reaction, a phase transition occurs between two components. The speed of the phase transition can be specified in the dimension of $\text{kg}/(\text{m}^3 \text{ s})$ and is calculated as follows:

$$\text{Mass Transfer} = \frac{\partial \alpha}{\partial t} \bar{\rho}. \quad (2.9)$$

Unfortunately, at this point a latent heat release during the phase transition is not implemented in ANSYS CFX, so it is necessary to imitate this effect in the form of additionally distributed heat source in the volume of the composite according to the law corresponding to the speed of the phase transition W/m^3 :

$$\text{Heat Source} = \frac{\partial \alpha}{\partial t} \rho_r H_u V_r. \quad (2.10)$$

The model of the phase transition in CFX has an additional technical limitation – it cannot be used when a contact is modelled between liquid and solid volumes. Therefore, for the problems where a calculation of the temperature field for the surrounding shapes is also required, the composite is modelled in another way – by introducing of additional scalar parameter equal to α with the intensity of the distributed source of this scalar corresponded to the *heatSource*. In this case the composite can also be modelled not as a uniform flow in the liquid volume but as a solid volume with a predetermined constant speed and including an advection at the input section.

The main advantage of the calculation in ANSYS CFX is that regardless of the method of implementation, the calculation changes of the degree of cure are done with a built-in method coupled with the calculation of thermodynamics and thus higher speed, accuracy and stability of the design scheme is achieved which is especially important when the degree of cure is very fast at some stage of the process. In addition, there is no need for a connection of the calculation time step with the size of the finite element in a pull direction. The main disadvantage of the procedure based on ANSYS CFX is the impossibility to use anisotropic thermal conductivities.

The developed procedures have been successfully validated by the temperature and degree of cure fields described in the literature studying the pultrusion of cylindrical rod [15], [21], [22], flat plate [5], [15] and I-beam profiles [16].

2.3. Curing Kinetic Modelling

For a simulation of the pultrusion process, the curing kinetic model for the matrix material should be developed. The resin curing process is an exothermic reaction. Heat generation depends on the resin absolute temperature and the degree of cure, which can be measured by a DSC (Differential Scanning Calorimetry) apparatus. There are two types of DSC test: dynamic and isothermal. In the isothermal DSC, the resin is kept in a constant temperature until the curing process completes. In the dynamic DSC, resin is heated with a constant rate (1 °C/min to 15 °C/min) [14]. In the pultrusion process, except near the wall of the die region, resin temperature rises almost at a constant rate [19]. So, in the present work the dynamic DSC test method has been applied to obtain the heat generation of the resin, which has been used for building of the curing kinetic model. A Microsoft Excel engineering tool for characterisation of the parameters of curing kinetic models has been developed.

In kinetic analysis, it is generally assumed that the rate of resin reaction can be described by Equation (2.3) consisting of two separable functions, the Arrhenius relationship $K(T)$, (Equation (2.4)) and the reaction function $f(\alpha)$, depending on the resin properties and varying with the applied resin reaction model. For the thermoset resins, traditional forms of the reaction function are

$$f(\alpha) = (1 - \alpha) - \text{first order kinetic model}, \quad (2.11)$$

$$f(\alpha) = (1 - \alpha)^n - n\text{-th order kinetic model}, \quad (2.12)$$

$$f(\alpha) = (1 - \alpha)^n (1 + K_2 \alpha) - n\text{-th order kinetic model with autocatalysis}, \quad (2.13)$$

$$f(\alpha) = \alpha^m (1 - \alpha)^n, \quad m + n = 2 - \text{Prout-Tompkins autocatalytic model}, \quad (2.14)$$

where m, n and K_i are empirical reaction constants.

In some cases two or even more sets of equations are used to describe the reaction. As a modified approximation for the rate of curing of thermoset resins, Kamal-Sourour autocatalytic model is more widely used

$$\frac{\partial \alpha}{\partial t} = \left(K_1 \exp\left(-\frac{E_1}{RT}\right) + K_2 \exp\left(-\frac{E_2}{RT}\right) \alpha^m \right) (1 - \alpha)^n. \quad (2.15)$$

Coefficients of the Arrhenius relationship, activation energy E and frequency factor K_0 , are physical values and they could be determined by the Kissinger method [12] or ASTM E 698 standard methodology [2] from the DSC dynamic tests. For a proper accuracy at least three DSC tests at different heating rates (usually 1 °C/min to 10 °C/min) are required.

In the case of the Kissinger method, the following equation is used:

$$\ln\left(\frac{\beta}{T_p^2}\right) = \ln\left(\frac{K_0 R}{E}\right) - \frac{E}{RT_p}, \quad (2.16)$$

where β is the heating rate, °C/s; and T_p is the peak temperature, °C.

By plotting $\ln(\beta / T_p^2)$ versus $1/T_p$, the values of activation energy E and frequency factor K_0 can be estimated by calculating the slope of the linear fit a_s and the y -intercept b_{int} . In this case the value of activation energy is determined as

$$E = -a_s R \quad (2.17)$$

and the value of frequency factor is determined as

$$K_0 = \frac{E e^{b_{int}}}{R}. \quad (2.18)$$

In the case of application of ASTM E 698 standard methodology, the plot of $\lg(\beta)$ versus $1/T_p$ is examined. Now the value of activation energy is determined as

$$E = -2.19 a_s R, \quad (2.19)$$

and the value of frequency factor is determined as

$$K_0 = \beta \frac{E e^{\frac{E}{RT_p}}}{RT_p^2}. \quad (2.20)$$

The coefficients of function $f(\alpha)$ are obtained by fitting of the experimental heat flow curves applying the least squares method:

$$\Delta = \sum_{i=1}^n \sum_{j=1}^m (\alpha_{ij}^{\text{exp}} - \alpha_{ij})^2 \rightarrow \min, \quad (2.21)$$

where n is the number of DSC tests at different heating rates; and m is the number of sampling points.

The developed Microsoft Excel tool for building of the curing kinetic models has been successfully validated by using DSC data obtained with Netzsch DSC-204 Phenix analyser for an epoxy resin in paper [1].

The parameters of Arrhenius relationship are obtained by the Kissinger method and ASTM E 698 procedure. The n -th order, Prout-Tompkins and Kamal-Sourour models have been chosen to fit the experimental heat flow curves.

The results of experimental curves' fitting are presented in Fig. 2.3 for the heating rate of 7.5 °C/min. It is necessary to note that the coefficients of Arrhenius relationship obtained by the Kissinger method have been used in the developed curing kinetic models.

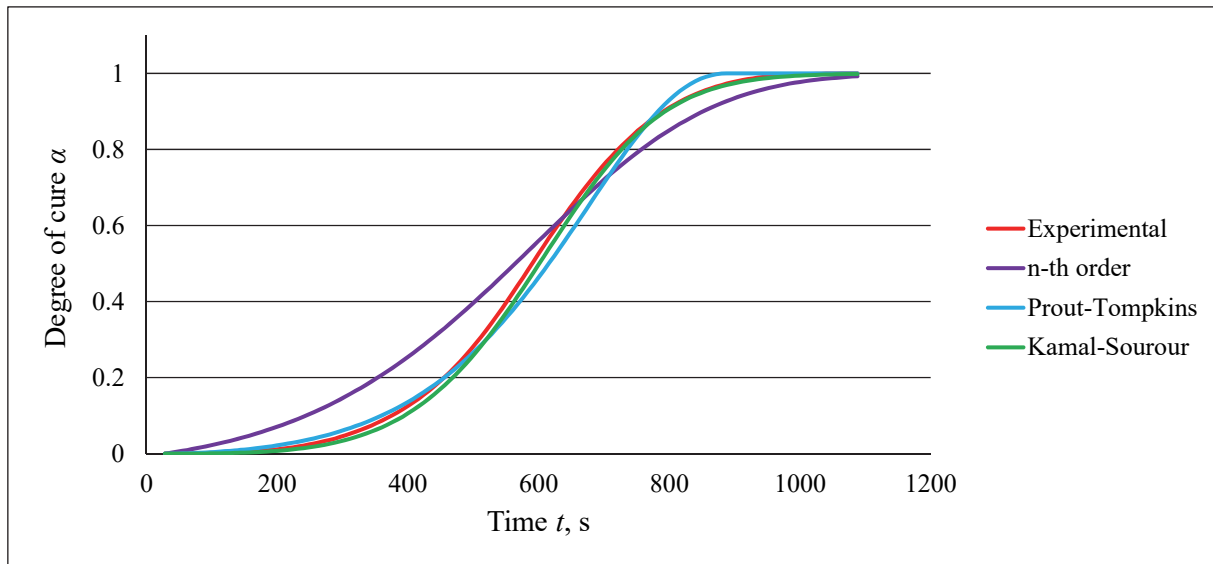


Fig. 2.3. Dependence of the degree of cure on time for the heating rate of 7.5 °C/min.

Fig. 2.3 clearly demonstrates that the best precision has been obtained with the Kamal-Sourour model but the n -th order model gave the worst result.

Finally the developed methodology has been successfully applied for building of the curing kinetic models of resin and coating with high microwave absorption properties to be used in advanced pultrusion processes. The following materials have been chosen for an application:

- coating: RESOLTECH RESOLCOAT 2010 FGCS (epoxy);
- resin: POLRES 305BV (polyester).

To define the curing kinetic parameters, results of DSC scans performed in AIMPLAS (Spain) by Perkin Elmer Diamond HeperDCS machine heating samples from 20 °C to 250 °C at heating rates of 2 °C/min, 5 °C/min, 10 °C/min are used. The parameters of Arrhenius relationship, activation energy and frequency factor, are determined by using the Kissinger method and, as the reaction function, the Kamal-Sourour model (Equation (2.15)) is used. Determined kinetic parameters (Tables 2.2 and 2.1) and experimental and modelled dependences of the degree of cure on temperature at different heating rates (Fig. 2.4) are given below for the coating and resin used in the investigation.

Table 2.1.

The determined parameters of curing kinetic model
of RESOLTECH RESOLCOAT 2010 FGCS

H_{tr} , J/kg	K_1 , s ⁻¹	K_2 , s ⁻¹	E_1 , J/mol	E_2 , J/mol)	n	m
152 907	2416 288	186 069 445 590 610	609 443	192 271	1.700	0.190

Table 2.2.

The determined parameters of curing kinetic model of POLRES 305BV

H_{tr} , J/kg	K_1 , s ⁻¹	K_2 , s ⁻¹	E_1 , J/mol	E_2 , J/mol)	n	m
323 074	14 289 310 986	285.870	85 573	33 141	2.342	0.519

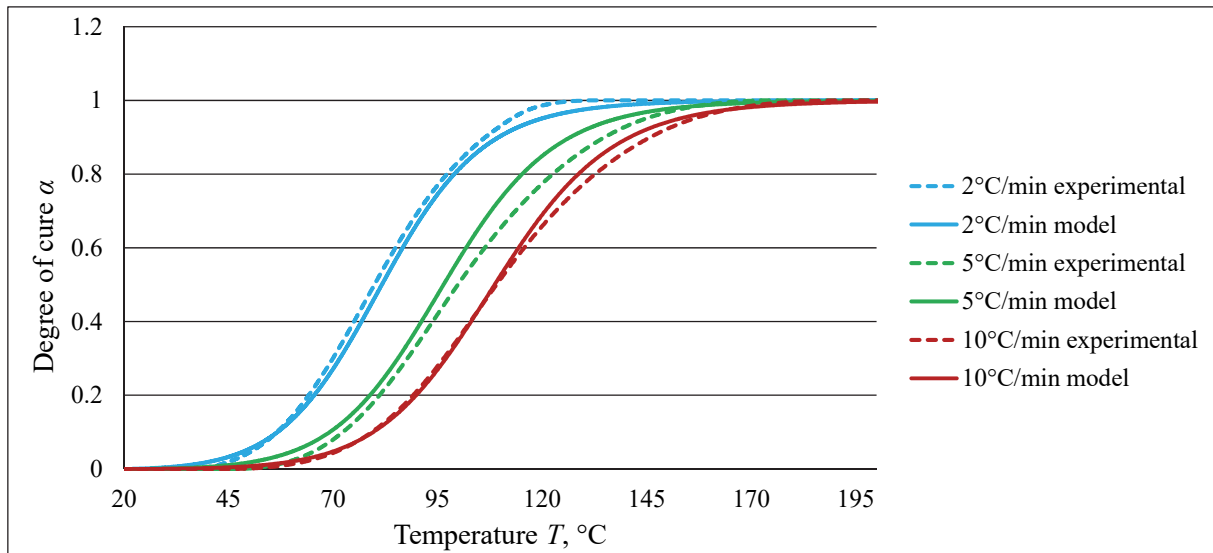


Fig. 2.4. Dependences of the degree of cure on temperature for RESOLTECH RESOLCOAT 2010 FGCS at different heating rates.

2.4. Experimental Validation of Simulation Algorithms

Conventional pultrusion experiment has been done in AIMPLAS (Spain). Experimental set-up and simplified model of the pultrusion tool is presented in Fig. 2.5. Heating of the die is realized by 12 electrical heaters split into three groups controlled by the proportional-integral derivative (PID) controller and thermocouples located between each group of heaters. Electrical power of each heater is 315 W. The controller turns off the heater groups when the temperature on the corresponding thermocouples reaches 100 °C, 120 °C or 140 °C (Table 2.3). Parameters of the pultrusion process, temperature and electrical resistance on the surface of the running pultruded profile, have been measured by three specifically designed sensors located on the die top and newly developed by Synthesites Innovative Technologies (Greece). The sensors are connected to the Optimold system [10] for data processing.

Pultrusion die has been made of steel 40Cr. The materials used for the production of the cylindrical rod are glass fibres Unifilo 4800 tex and polyester resin POLRES 305BV. The fibre volume content in the pultruded material is 55 %. The rate of resin reaction is described by using the Kamal-Sourour curing kinetic model (Equation (2.15)) developed in Chapter 3. The experimental pultrusion process is realized at pull speed of 18 cm/min. Room temperature is 17 °C, resin temperature is equal to the room temperature since no resin preheating is used.

Table 2.3

Thermocouple	Control temperatures			
	Initial, °C	Corrected 1, °C	Corrected 2, °C First switch- off	Main control
1	100	85	65	85
2	120	105	85	105
3	140	125	105	125

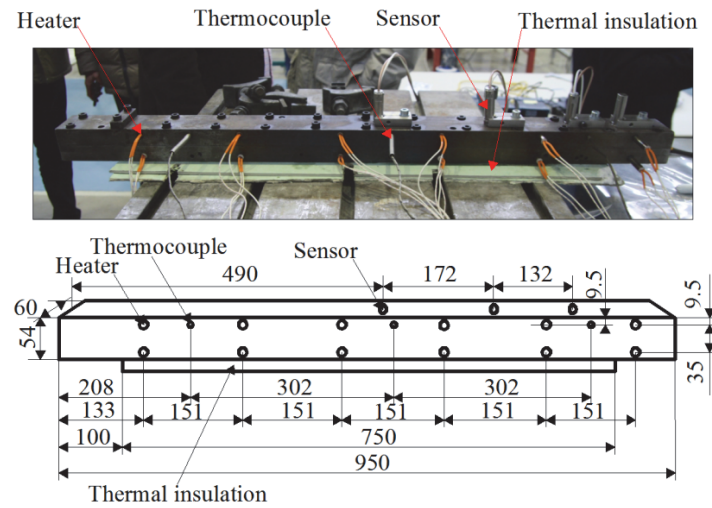


Fig. 2.5. Set-up of conventional pultrusion.

The first finite element model for a simulation of the cylindrical rod pultrusion has been created in ANSYS Mechanical by using 3-D thermal solid finite elements Solid 70. In this case symmetry of the simulated domain is used and only a quarter of the die is modelled. To analyse the heat transfer and curing processes in the post-die region, the modelling of the profile is continued at the distance of 250 mm from the die exit. A fragment of the finite element model is presented in Fig. 2.6a.

The results of simulation, temperature and degree of cure obtained on Sensor 3, together with experimental measurements are presented in Fig. 2.7. It demonstrates that close agreement between experimental and simulation results has not been obtained. If to suppose that experimental results are correct, a disagreement should be identified.

The following corrections have been done to minimize the difference between numerical and experimental results:

- correction of control temperatures for heater groups (Table 2.3, Corrected 1);
- correction of temperature control algorithm (Table 2.3, Corrected 2);
- correction of die material thermal properties.

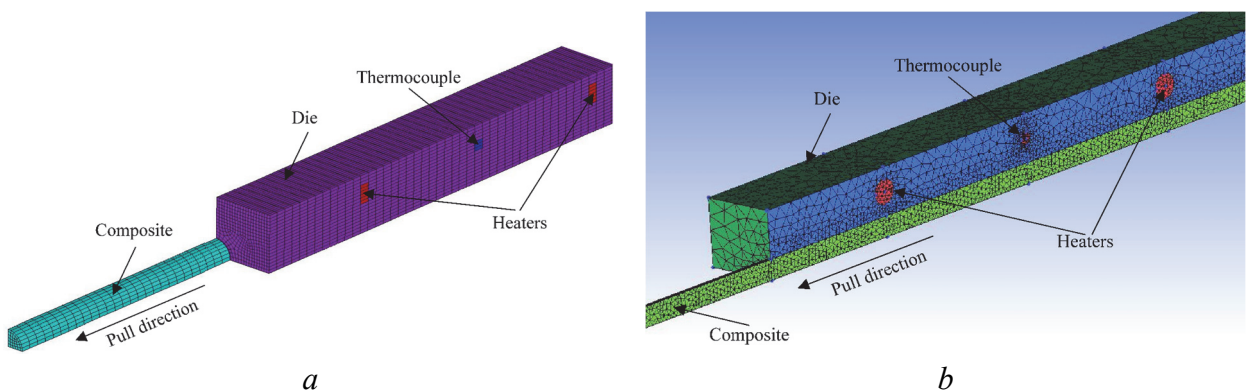


Fig. 2.6. Fragment of the finite element models: *a* – ANSYS Mechanical, *b* – ANSYS CFX.

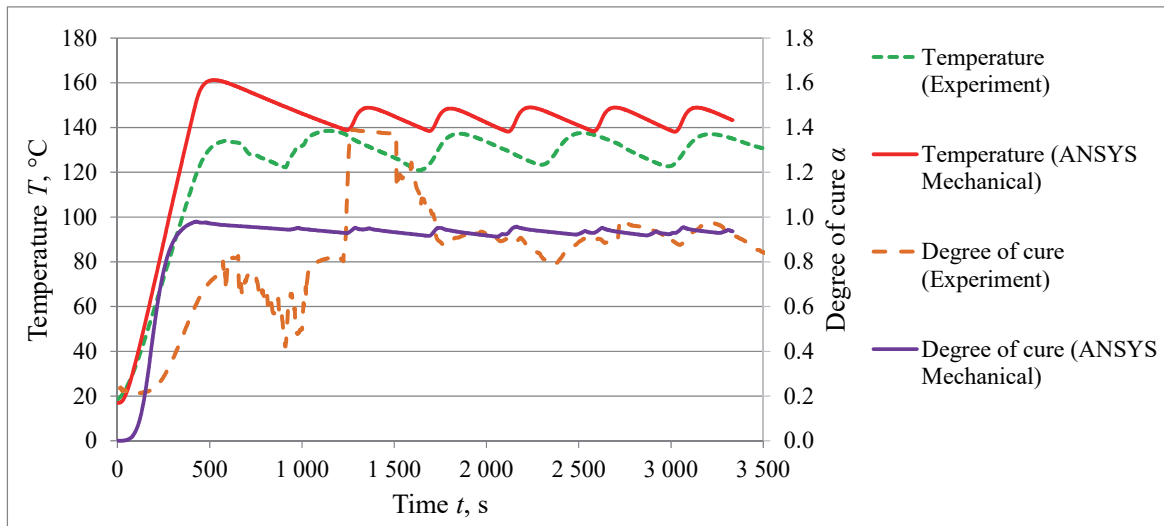


Fig. 2.7. Temperature and degree of cure on Sensor 3.

Good agreement between experimental and simulation results has been obtained after these corrections (Fig. 2.8).

To validate and evaluate the ANSYS CFX based calculation algorithm, the problem in the final formulation has been solved in ANSYS CFX. The fragment of the FE model is presented in Fig. 2.6b. Due to CFX software limitations, an isotropic thermal conductivity as lumped composite material property had to be considered with the average value of k_x and k_y . The results of this simulation together with experiment results and results obtained by ANSYS Mechanical can be seen in Fig. 2.8. Slight difference is observed in temperature and degree of cure curves obtained by ANSYS Mechanical and ANSYS CFX.

To ensure that this difference is caused by different pultruded material thermal conductivity used in the simulation and is not connected with the simulation algorithm, additional simulation has been performed in ANSYS Mechanical using averaged lumped thermal conductivity of profile material used in ANSYS CFX. The results of this simulation are shown in Fig. 2.9. Now it is clear that the difference obtained previously is connected only with the different material properties used in the simulations.

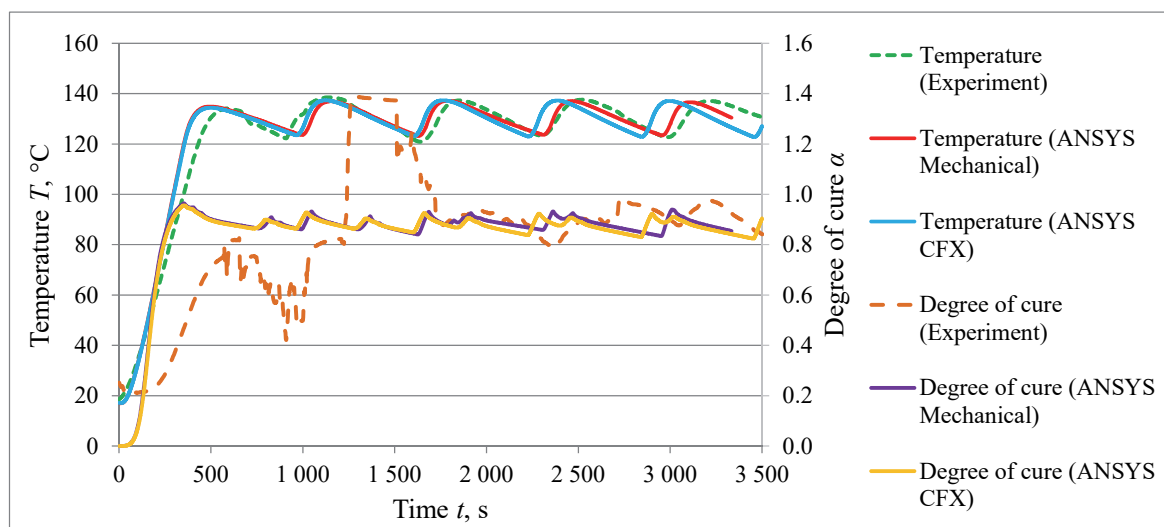


Fig. 2.8. Temperature and degree of cure on Sensor 3 obtained by ANSYS Mechanical and ANSYS CFX.

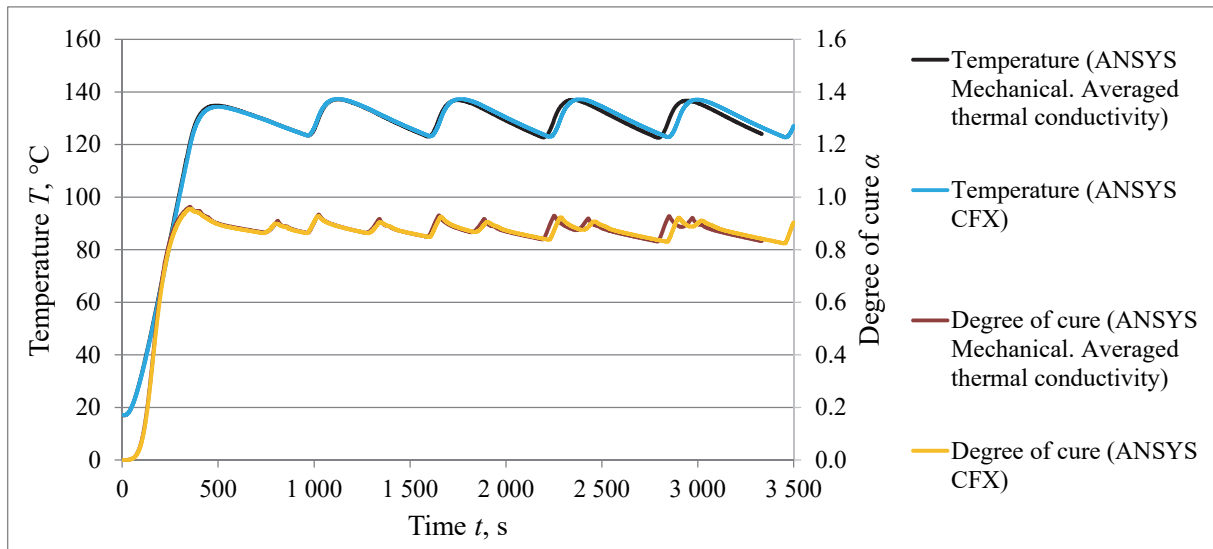


Fig. 2.9. Temperature and degree of cure on Sensor 3 obtained by ANSYS Mechanical and ANSYS CFX.

2.5. Modelling of Microwave Assisted Pultrusion Processes

New simulation methodology consisting of two sub-models is developed for simulation of the microwave assisted pultrusion processes. Each of the sub-models is constructed by using the general-purpose FE software. In the first step the electromagnetic sub-model is used to evaluate the electric field distribution by using the COMSOL Multiphysics software. In the second step an absorption energy field obtained in the composite material is applied as a heating source in the thermochemical sub-model developed in ANSYS Mechanical environment to determine the temperature and degree of cure fields in the pultruded composite. To demonstrate the application of the developed methodology for the design of technological process, the microwave assisted pultrusion of the cylindrical rod is investigated.

The electromagnetic problem is solved with the purpose to determine the electric field distribution and as the result to obtain the absorbed energy field in the composite material.

Using the common approach of a harmonic oscillating electric field \vec{E} ,

$$\vec{E}(\vec{r}, t) = \vec{E}(\vec{r}) e^{2\pi i f t}, \quad (2.22)$$

where \vec{r} is the location vector; t is a time, s; and f is the microwave frequency, Hz.

Maxwell's equations could be written as follows:

$$\nabla \times \nabla \times \vec{E}(\vec{r}) - \varepsilon_0 \mu_0 (2\pi f)^2 \varepsilon_r \vec{E}(\vec{r}) = \vec{0}, \quad (2.23)$$

where ε_0 is the vacuum permittivity, F/m; and μ_0 is the magnetic constant, H/m.

This complex valued equation is solved numerically for the amplitudes of the electric field $\vec{E}(\vec{r})$ with respect to the relative permittivity ε_r , which for loss dielectric materials like mixtures of fibers and resin is a complex function of frequency f , temperature T and degree of cure α :

$$\varepsilon_r(f, T, \alpha) = \varepsilon'(f, T, \alpha) - i\varepsilon''(f, T, \alpha). \quad (2.24)$$

It can be obtained by the cavity perturbation method [20] at fixed frequencies or Corbino probe measurements [6], [7] with variable frequency. After numerical solution of Equation (2.23), the absorbed microwave energy generated by dielectric losses could be obtained as described in [18]:

$$Q(\vec{r}) = 2\pi f \varepsilon_0 \varepsilon'' |\vec{E}(\vec{r})|^2. \quad (2.25)$$

Knowing the absorbed energy field Q in the composite material, the thermochemical problem described and numerically solved in Chapter 2 can be used.

To demonstrate the application of the developed methodology for the design of technological process, the microwave assisted pultrusion of the cylindrical rod with the diameter of 16 mm and made of glass fibers Unifilo 4800 tex and polyester resin POLRES 305BV has been investigated.

In the present case, the base model has to provide a symmetric cylindrical field distribution with an extended maximum of electrical field covering the product diameter. The centrepiece of the microwave system consists of a ceramic part (ZrO_2) with a 16 mm hole, which has been inserted as a section into the metal pultrusion die. The objective was to achieve that the microwave field inside the hole and therefore inside the rod running through that hole is as homogenous as possible. A magnetron of frequency 915 MHz and a waveguide WR 975 have been used.

Fig. 2.10 shows the scheme of the electromagnetic sub-model for microwave pultrusion with the ceramic inlet and coupling of the microwave to the ceramic part with a waveguide.

Fig. 2.11 shows the distribution of the resulting heating rate Q in different 3D- and cross-sectional views for the optimized geometry of the ceramic inlay. In Figs. 2.11(a) and 2.11(b) it is obvious, that only the rod inside the ceramic inlet is heated. The microwave coupling and the pultrusion die are not heated by the microwave. Outside of the ceramic inlet the microwave field is close to zero. Fig. 2.11(c) shows the cross-section of the heating rate in the die and the waveguide. The heating rate is homogeneously concentrated in the rod. Additionally a small heating effect inside the ceramic material itself gets visible.

The advanced pultrusion die consists of microwave block with the ceramic inlet located at the entrance of the steel die (Fig. 2.12). The fragment of corresponding finite element model created in ANSYS Mechanical is presented in Fig. 2.13. Heating of the composite profile is realized by using an absorption energy field obtained by COMSOL Multiphysics. It is simulated only in the composite material within the boundaries of the ceramic inlet. The simulated absorption energy field is shown in Fig. 2.14. It is necessary to note that temperature control is not applied in this case and energy source is working continuously. Parameters of the pultrusion process are presented in Table 2.4.

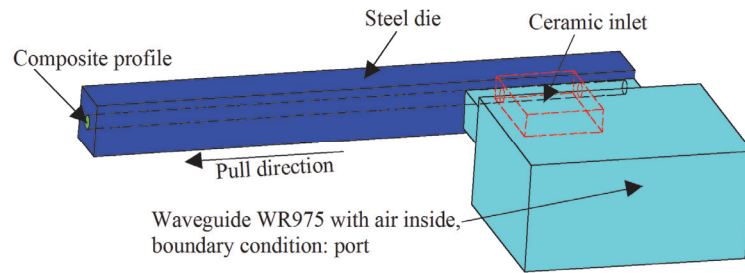


Fig. 2.10. Scheme of the electromagnetic model.

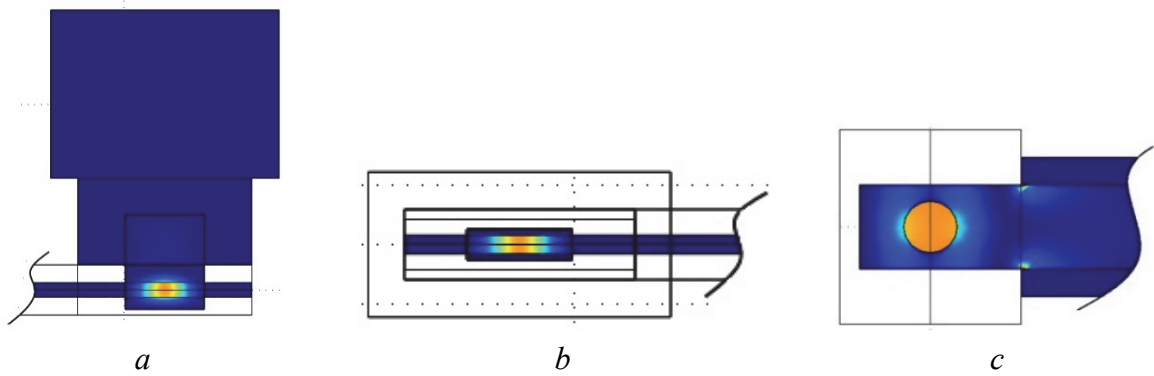


Fig. 2.11. Distribution of the heating rate Q : a – top-view, a – side-view and c – cross-section.

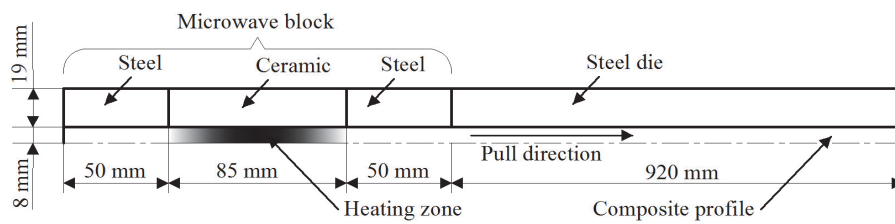


Fig. 2.12. Scheme of the advanced pultrusion die.

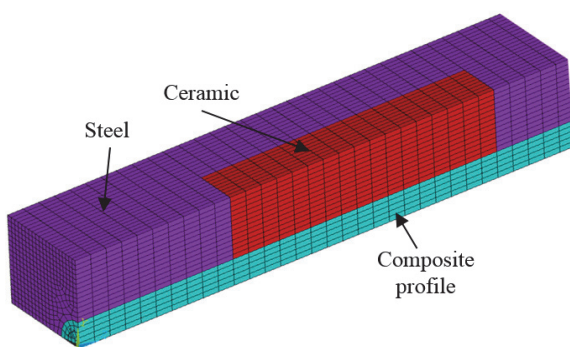


Fig. 2.13. Fragment of the finite element model.

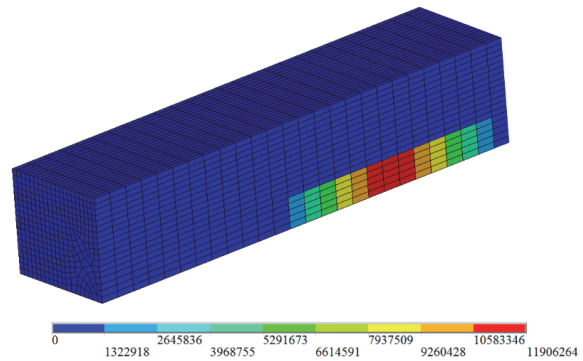


Fig. 2.14. Simulation of the absorption energy in the pultruded profile.

Table 2.4.

Parameters of pultrusion process	
Name, unit	Value
Room and resin temperature in the bath, °C	17 or 27 or 37
Pulling speed, m/min	0.18 or 0.12 or 0.06

The rate of resin reaction is described by using the Kamal-Sourour curing kinetic model (Equation (2.15)) with the parameters presented in Table 2.2. To increase the microwave energy absorption properties of the resin a filler Martinal of mass content of 20 % is added to the resin.

Results of simulation, temperature and degree of cure at the profile centreline and surface, are given for the time 50 min from starting of the pultrusion process in Fig. 2.15 at room temperature 27 °C and pull speed 12 cm/min. It can be seen that, as opposed to conventional pultrusion process, the curing now comes considerably faster in the profile centreline than on the surface. Thus high degree of cure in the profile centreline is reached at the beginning of steel die already for the pultrusion process with pull speed equal to 18 cm/min and room temperature equal to 37 °C. Moreover, full curing in the profile centreline happens already in the ceramic inlet for the pultrusion processes with pull speed equal to 6 cm/min and at all examined room temperatures while the degree of cure at the profile surface has not reached high values.

To obtain the high value of the degree of cure at the profile surface in the advanced pultrusion die and to increase the pull speed of the process, thus improving the effectiveness of the pultrusion process, an influence of the following parameters of pultrusion process has been investigated:

- reduction in twice of steel die length;
- application of thermal insulation at all external surfaces of advanced pultrusion die (both steel die and microwave block);
- preheating of steel die, excluding microwave block, at the uniform temperature of 60 °C before the beginning of pultrusion process;
- heating of resin bath (composite) at the temperature of 50 °C for full duration of pultrusion process.

The influence of the examined factors has been studied for the pultrusion process with the room temperature of 27 °C and pulling speed of 12 cm/min. It was shown that the reduction of steel die length has no effect on the degree of cure at the profile surface. However, any reduction of all important parameters has not been observed either. It demonstrated that an application of thermal insulation at all external surfaces of advanced pultrusion die has by 36 % increased the value of the degree of cure at the profile surface. Also, it has been shown that only by preheating of steel die with the temperature of 60 °C before the beginning of pultrusion process, it is possible to increase the value of the degree of cure at the profile surface by 29 %. It was demonstrated that heating of resin bath had by 44 % increased the value of the degree of cure at the profile surface that could be used also in the design of advanced pultrusion die. However, it is necessary to note that the effectiveness of the pultrusion process could not be so high since the heating of resin bath was realized for the full duration of pultrusion process.

Additionally, joint influence of two factors having large effect on the degree of cure at the profile surface and on the process effectiveness, namely an application of thermal insulation and preheating of steel die, has been investigated. It was shown that by preheating of steel die at the temperature of 60 °C before the beginning of pultrusion process and applying the thermal insulation, it is possible to increase the value of the degree of cure at the profile surface by 79 %. It is necessary to note that joint influence of the two examined factors (79 %) is higher than their separate influence summarized together (65 %).

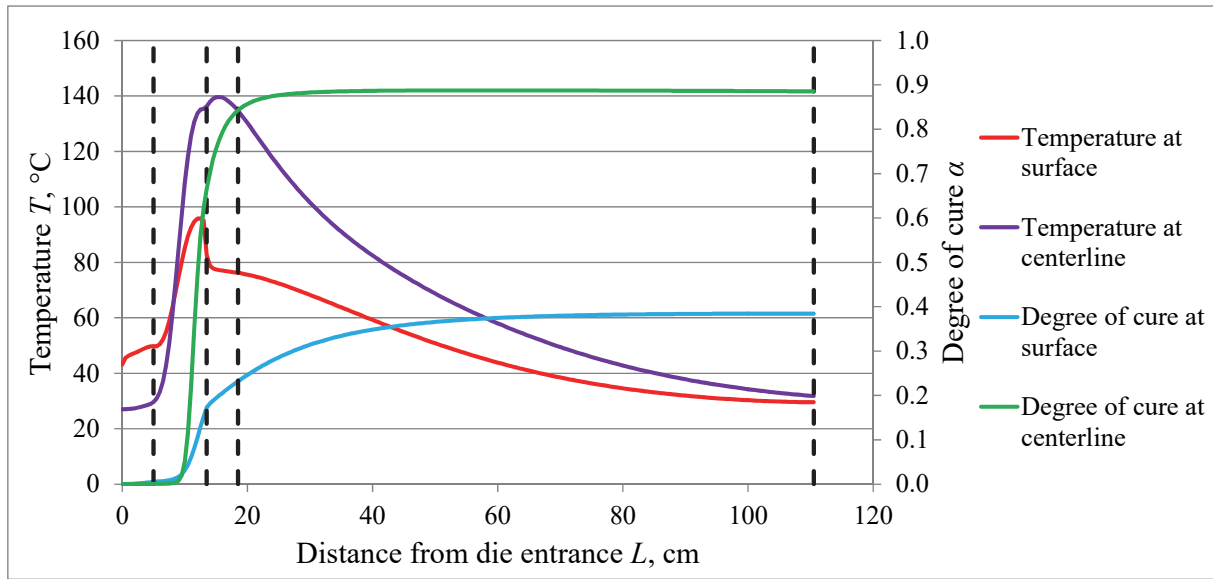


Fig. 2.15. Temperature and degree of cure at the profile centerline and surface at room temperature 27 °C and pull speed 12 cm/min.

To obtain the highest values of the degree of cure at the profile surface in the advanced pultrusion die, an influence of the capacity of the absorption energy field on the parameters of pultrusion process has been investigated. It is necessary to note that the distribution of the absorbed energy in the composite profile is not changed while its intensity has been increased proportionally with the coefficients 1.5, 2.0 and 2.5. Fig. 2.16 shows that the high value of the degree of cure at the profile surface could be reached already in the ceramic inlet of the microwave block. However, it is necessary to note that the material temperature in this case could be higher than allowable resin temperature. To avoid overheating of the resin, the pulling speed should be increased at the same time contributing to the considerable increase of effectiveness of the microwave assisted pultrusion process.

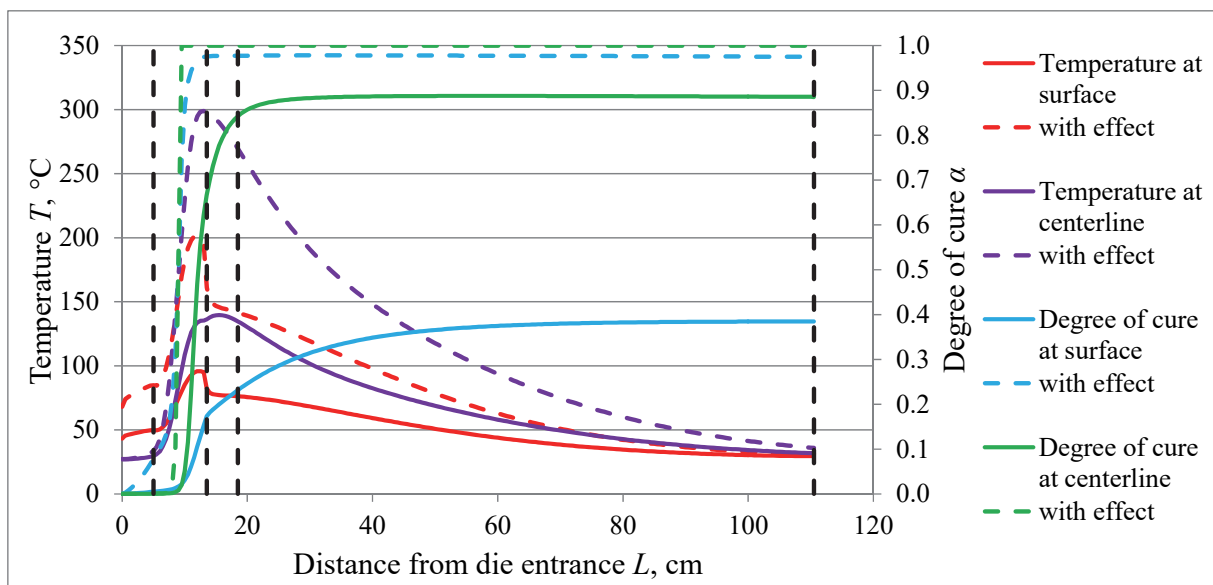


Fig. 2.16. Temperature and degree of cure at the profile centreline and surface with the intensity coefficient of the absorption energy field equal to 2.5 (pull speed: 12 cm/min).

2.6. Design of Multifunctional Pultrusion Die

The main idea of multifunctional die is to combine profile curing and application of the coating in one pultrusion die. This advanced process allows reducing the number of manufacturing steps (Figs. 1.2 and. 1.3). This process is free of VOCs and small particle emission. Also, the proposed process reduces labour and process costs. One more benefit is the extraordinary interaction between coating and profile achieved by the cure of the coating over a non-fully cured profile. Profile resin degree of cure in the range from 0.75 to 0.85 is required for the best interaction between profile and coating. The scheme of multifunctional pultrusion die is given in Fig. 1.4. After the application of coating, the profile resin must be fully cured together with coating.

Since the profile resin is cured by microwaves, the profile is heated from centre to surface and curing reaction also starts from the centre. Curing reaction is exothermic, therefore theoretically it is possible to cure the profile surface and coating by exothermic heat accumulated in profile. The coating injection chamber should be placed as close as possible to the microwave block, because the temperature of profile material rises precipitously after the heating zone.

The possibility to cure the coating by the heat generated by microwaves and exothermic profile resin reaction is estimated for the pull speed of 18 cm/min, room temperature of 27 °C and applied microwave energy of 2.35 kW. The coating is applied at the distance of 100 mm after the microwave block (Fig. 2.17). Coating material is epoxy RESOLTECH RESOLCOAT 2010 FGCS coat. Coating layer thickness is 0.5 mm. Before the injection the coating is preheated to the temperature of 50 °C.

The results of this simulation, temperature and degree of cure at the profile centreline and surface and at the coating inner, middle and outer surfaces are given for the time of 50 min from starting the pultrusion process in Fig. 2.18.

High degree of cure ($\alpha \geq 0.95$) in the profile centreline is reached already at the beginning of steel die right after the microwave block. The degree of cure at the profile surface after the microwave block is equal to the desired value 0.8 (for a good adhesion with coating). It is shown in Fig. 2.18 that the applied coating is not completely cured. The degree of cure of the coating on the profile surface is equal to 0.31 but the degree of cure of the coating on its outer surface is 0.24.

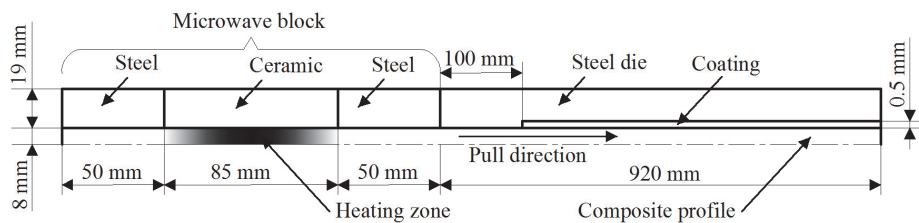


Fig. 2.17. Simulation domain.

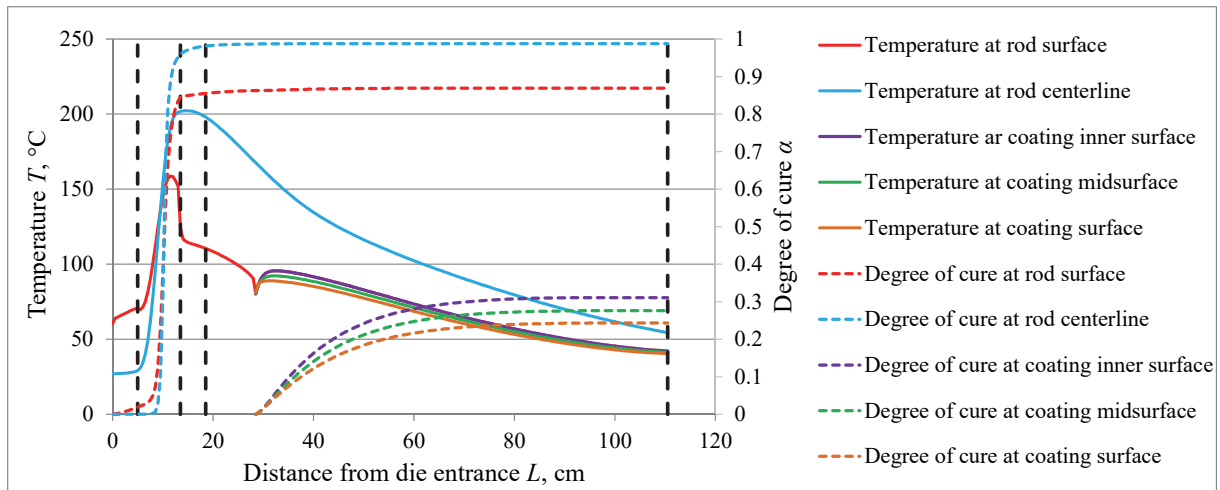


Fig. 2.18. Temperature and degree of cure in the profile and coating.

To increase the degree of cure of coating, the influence of the following parameters of pultrusion process has been investigated:

- application of thermal insulation at all external surfaces of advanced pultrusion tool (both steel die and microwave block);
- preheating of steel die with the uniform temperature of 60 °C before the beginning of pultrusion process;
- joint application of thermal insulation and preheating of the die;

It has been shown that thermal insulation and preheating of the pultrusion tool increases the degree of cure in the coating, but does not contribute to its complete curing (Fig. 2.19). Also, it is important to note that when the thermal insulation is applied, the degree of cure of the profile surface increases from 0.85 to 0.90 therefore the applied energy could be a little bit decreased. The decrease of the applied microwave energy also is required because of the profile resin overheat (the maximal temperature is about 210 °C and the allowable is 190 °C).

All these results show that it is impossible to obtain the complete curing of the applied coating only by the microwave heating therefore some electrical heaters should be applied in the pultrusion process.

Two electrical heaters with power of 315 W placed at the distance of 100 mm from the coating injection point and controlled at the temperature of 160 °C have been introduced to the multifunctional pultrusion die to evaluate the effectiveness of electric heaters for secondary heating of the pultruded profile. The pull speed and room temperature is the same as in the previous simulations ($V = 18$ cm/min, $T_{\text{room}} = 27$ °C), but the applied microwave energy is decreased to 1.9 kW to avoid the resin overheat. The scheme of the simulation domain is shown in Fig. 2.20. The results of this simulation for the time of 50 min after starting the process are shown in Fig. 2.21.

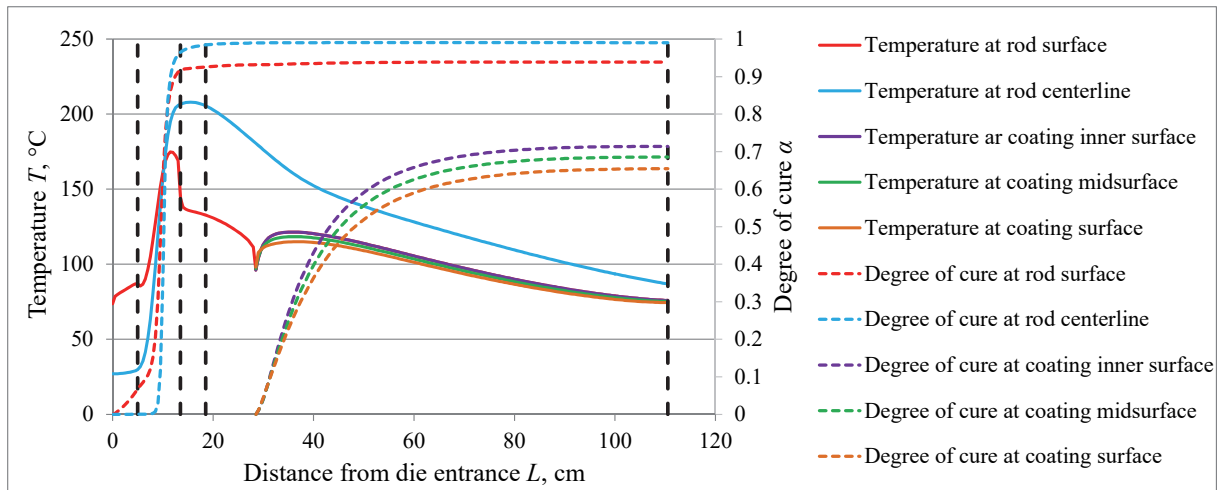


Fig. 2.19. Temperature and degree of cure in the profile and coating taking into account both effects of thermal insulation and preheating of the pultrusion tool.

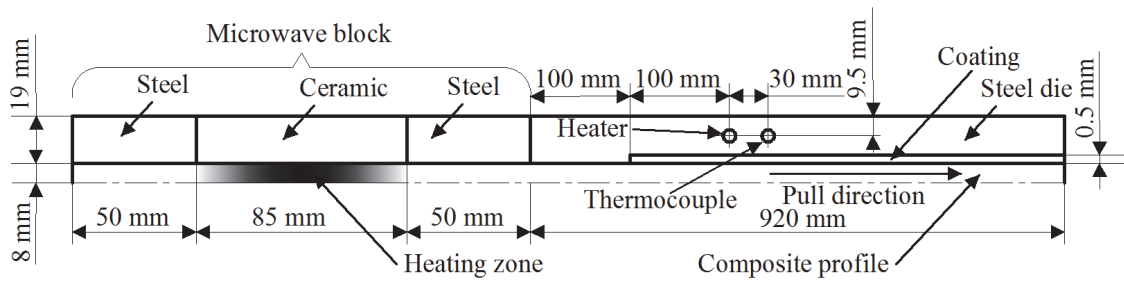


Fig. 2.20. Simulation domain.

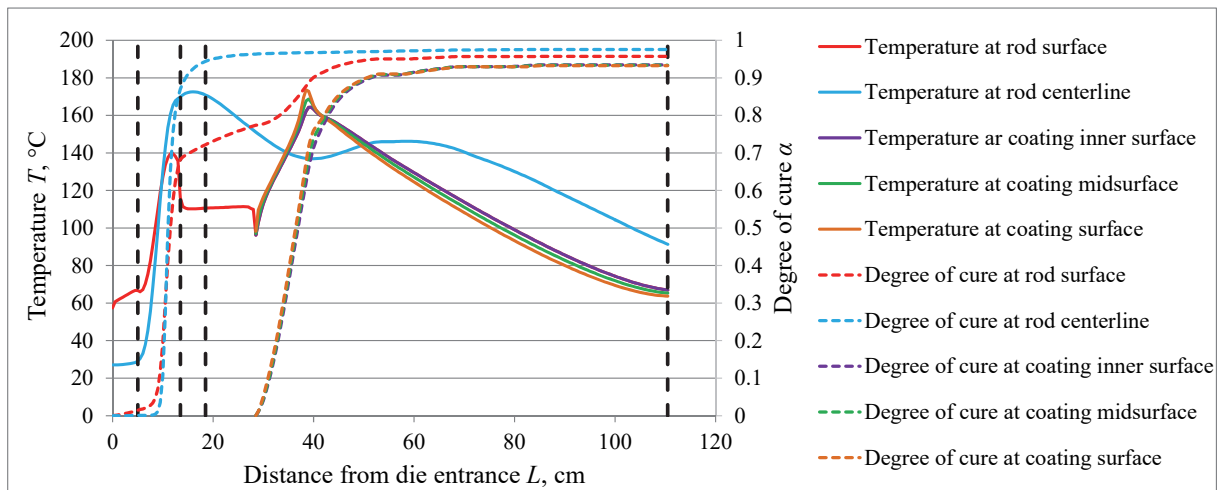


Fig. 2.21. Temperature and degree of cure in the profile and coating. Heating: MW and 2 electric heaters. Control temperature: 160 °C.

The profile and coating resins are not overheated now. The resin at profile centerline is almost completely cured already in the microwave block. The degree of cure at profile surface at the coating injection point is 0.78 that is required for good adhesion between the profile and coating. Electric heaters produce enough energy to cure the profile resin at the surface ($\alpha = 0.96$) and the coating ($\alpha = 0.93$). The results of simulation clearly show that it is possible to obtain completely cured coated profile.

2.7. Optimisation of Microwave Assisted Pultrusion Process for Multifunctional Die

Due to the large dimension of numerical problems to be solved, an optimisation methodology is developed employing the method of experimental design [24] and response surface technique [17]. This methodology is a collection of mathematical and statistical techniques that are useful for the modelling and analysis of problems in which a response of interest is influenced by several variables and the objective is to optimise this response. An engineering approach of optimisation based on experimental design and response surface technique is presented in Fig. 2.22.

The optimisation of pultrusion process parameters is executed for the rod profile with the diameter of 16 mm made of fiberglass Unfilo 4800 tex (mass content of 70 %) and polyester resin POLRES 305BV. To increase the microwave energy absorption properties for the resin, filler Martinal with mass content of 20 % is added to the resin. The profile is coated by epoxy coating RESOLTECH RESOLCOAT 2010 FGCS in the same pultrusion die. The thickness of coating layer is 0.5 mm.

Fig. 2.23 presents CAD drawing of the pultrusion tool. The die is made of steel 40 Cr. Dry glass fibers come into the die and are saturated with the resin in the resin chamber making the composite profile. The resin is partly cured by the microwave energy to obtain the degree of cure $\alpha = 0.75$ to 0.85 on the profile surface required for a good adhesion between the profile and coating. Then it is coming through the coating chamber where the coating material is applied. Then the coating and profile are heated by 8 electrical heaters split into 2 groups and controlled by PID controller and thermocouples located near heaters. The electrical power of each heater is 315 W. Three holes are provided in the die for an installation of Optimold monitoring system [10] sensors, which monitor the temperature and the degree of cure on the surface of running profile in real time.

The scheme of the simulation domain is given in Fig. 2.24. Corresponding finite element model presented in Fig. 2.25 has been created in ANSYS Mechanical by using 3-D thermal solid finite elements Solid 70. Using the symmetry of the simulated domain, only a quarter of advanced pultrusion die is modelled.

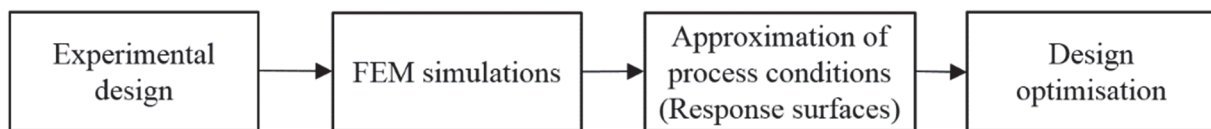


Fig. 2.22. Optimisation process.

where W_{mw} is applied microwave power, kW; W_{heater} is power of electrical heater, kW; n is number of electrical heaters; V_{pull} is pull speed, m/h; and k_t is relative time of heaters work during the simulation time:

$$k_t = \frac{T_{work}}{T_{sim}}, \quad (2.27)$$

where T_{work} is time of heaters work, h; and T_{sim} is total simulation time, h.

Some parameters of the process, namely, the number and power of electrical heaters and coating preheating temperature are fixed. Their values are given in Table 2.5. Upper and lower bounds (domain of interest) for the variable parameters of the technological process are also listed in Table 2.5.

Constrains are introduced into optimisation procedure with the aim to provide qualitative profile production when profile and coating resins should be completely cured at die exit and the materials should not be overheated during the pultrusion process. Constrains of the process are summarized in Table 2.6.

Table 2.5.

Constant and variable parameters of pultrusion process			
	constant	min value	max value
Coating preheating temperature, °C	50	—	—
Power of electrical heater, W	315	—	—
Number of electrical heaters	8	—	—
Pull speed, cm/min	—	30	100
Room and resin temperature, °C	—	12	40
Applied microwave energy, kW	—	2	6
Control temperature of electrical heater, °C	—	70	150

Table 2.6.

Constrains of pultrusion process		
	min value	max value
Temperature in profile, °C	—	190
Temperature in coating, °C	—	250
Degree of cure at die exit	0.93	—
Degree of cure at coating injection point	0.75	0.85

Table 2.7.

Standard deviations and relative errors of approximations					
Symbol, unit	Approximation with all sampling points		Number of eliminated points	Approximation after elimination of some points	
	σ_{err}^0	Δ , %		σ_{err}^0	Δ_1 , %
T_{cent} , °C	8.898	15.8	2	5.5160	6.2
α_{cent}	0.0929	479.7	6	0.0349	8.2
T_{surf} , °C	5.446	8.7	0	—	—
α_{surf}	0.0372	16.2	6	0.0317	10.1
$\alpha_{\text{coat inject}}$	0.0826	1955.9	8	0.0164	10.1
T_{coat} , °C	1.822	2.4	0	—	—
α_{coat}	0.0230	34.2	7	0.0128	4.9
k_t	0.00301	10.0	0	—	—

It is well known that, if for global approximations it is planned to use the second order polynomial functions, the D-Optimal experimental design is the most suitable [4]. The minimal number of design experiments in the optimisation problem is determined in this case as $n = (k+1)(k+2)/2$ where k is the number of design parameters. It is common practice to use two times greater number of points, therefore 30 sampling points have been used. D-Optimal plan of experiments has been generated by EDAOpt software [3].

After finite element calculations in each of 30 sampling points, the second order polynomial functions are obtained for simulation results using the conventional unweighted least square method. As stated in [17], there is a considerable practical experience indicating that second-order models work well in solving real response surface problems.

The following behaviour functions have been obtained: the maximal temperature of coating (T_{coat}), profile surface (T_{surf}) and centreline (T_{cent}), the degree of cure at die exit for coating (α_{coat}), profile surface (α_{surf}) and centreline (α_{cent}), and on the profile surface for coating injection point ($\alpha_{\text{coat inject}}$), and the relative time of heaters work (k_t). It is necessary to note that only approximation of k_t directly enters into the objective function (Equation (2.26)). Approximation of temperatures and degrees of cure are used as constrains during the optimisation procedure.

The standard deviations σ_{err}^0 and maximal relative errors Δ of approximation functions obtained using all sampling points are presented in Table 2.7. It is seen from Table 2.7 that relative errors of approximations are quite large (column Δ) especially for the points with low temperature and degree of cure for a composite material. Since these points are out of interest, it is possible to exclude them from approximations thus increasing their accuracy and narrowing the boundaries of the design space. It is necessary to note that no more than 8 points have been excluded from the examined design space. There are points with values lower than 0.25 for the degree of cure and points with values lower than 60 °C for the maximal temperature in composite material. The relative errors of new approximation functions are given in column Δ_1 of Table 2.7. It is shown that elimination of some points located out of interest have given the possibility to significantly improve the accuracy of approximations. The maximal relative error of approximations does not exceed 10 % now.

Taking into account the constraints presented by approximation functions, the minimum of the objective function (Equation (2.26)) has been found by two methods: new version of the random search (RS) method [11] using EDAOpt optimisation software [3] and the generalized reduced gradient (GRG) algorithm [13] utilized in Microsoft Excel.

The results of minimisation of energy consumption by the GRG method are presented in Fig. 2.26. Appropriate results obtained by RS method are presented in Fig. 2.27. No visible difference is observed in the results obtained by different methods. Since the response surfaces are used instead of the original functions, the optimal result is checked using ANSYS Mechanical finite element solution. Good coincidence of results is observed (Table 2.8).

As it is seen from Figs 2.26 and 2.27, the profile material is fully cured (degree of cure more than 0.98), but the coating material is not fully cured (degree of cure 0.93). At the same time the heaters control temperature comes to the upper border of the domain of interest (150 °C), but the pull speed – to the lower one (30 cm/min). Since the degree of cure of coating depends on the pull speed and temperature on the heaters, practically it is not possible to obtain its higher value without changing the corresponding domains of interest in optimisation problem. Taking into account that the maximal coating temperature (158.4 °C) is much less than the allowed temperature (250 °C) and the maximal temperature on profile surface (163.2 °C) is also less than the allowed temperature (190 °C), it is necessary to increase the heater control temperature that is technologically possible to be done.

This way the maximal control temperature of heaters work has been increased from 150 °C to 170 °C. To exclude the development of a new plan of experiments and time-consuming finite element calculations in each point of the plan, an extrapolation of approximations has been done as presented in Fig. 2.28. It is necessary to note that the values of the degree of cure are limited by 1.0. The results of optimisation with modified design space are shown in Fig. 2.29.

	Pull speed, cm/min	Room and resin temperature, °C	MW Power, kW	Heaters control temperature, °C	Maximal temperature at centerline, °C	Degree of cure at centerline	Maximal temperature at profile surface, °C	Degree of cure at profile surface	Degree of cure on profile surface at coating injection point	Maximal temperature in coating, °C	Degree of cure in coating	Relative time of heaters work k_t	Energy consumption, kWh/m
Optimal	31.0	21.3	3.42	150.0	185.68	0.99	163.15	0.98	0.75	158.43	0.93	0.139	0.203
min	30	12	2	70		0.93		0.93	0.75		0.93		
max	100	40	6	150	190		190		0.85	250			

Fig. 2.26. Optimal results obtained by the GRG method.

Optimization: COALINE_elimination.prj													
Criterion: $1.6667 \times 10^{-3} + 0.315 \times 8 \times Y_8 \times X_1$										= 0.202749993509009			
Indices No	Min	Type	Max	Criterion=	0.20274999								
1) X1:	30	0	100	1) X1=	30.656118								
2) X2:	12	0	40	2) X2=	22.015749								
3) X3:	2	0	6	3) X3=	3.3816248								
4) X4:	70	0	150	4) X4=	150								
5) Y1:	0	2	190	5) Y1=	186.16949								
6) Y2:	0.93	1	2	6) Y2=	0.99593112								
7) Y3:	0	1	190	7) Y3=	163.64049								
8) Y4:	0.93	1	2	8) Y4=	0.97614721								
9) Y5:	0.75	1	0.85	9) Y5=	0.75000132								
10) Y6:	0	1	250	10) Y6=	158.5069								
11) Y7:	0.93	1	2	11) Y7=	0.93000024								
12) Y8:	0	0	1	12) Y8=	0.13794337								

Fig. 2.27. Optimal results obtained by the RS method.

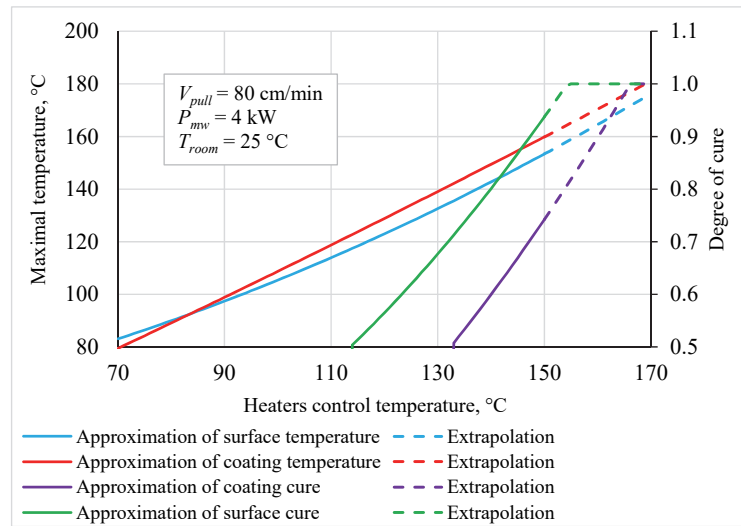


Fig. 2.28. Approximated and extrapolated dependences of temperature and degree of cure on heaters control temperature.

	Pull speed, cm/min	Room and resin temperature, °C	MW Power, kW	Heaters control temperature, °C	Maximal temperature at centerline, °C	Degree of cure at centerline	Maximal temperature at profile surface, °C	Degree of cure at profile surface	Degree of cure on profile surface at coating injection point	Maximal temperature in coating, °C	Degree of cure in coating	Relative time of heaters work k_t	Energy consumption, kWh/m
Optimal	63.9	40.0	4.98	169.2	165.33	0.93	166.38	1.00	0.75	176.24	1.00	0.153	0.140
min	30	12	2	70		0.93		0.93	0.75		0.93		
max	100	40	6	170	190		190		0.85	250			

Fig. 2.29. Optimal results obtained by GRG method for the modified design space.

The optimal values of design parameters together with corresponding values of constrains and objective function obtained by approximations and validated by ANSYS Mechanical simulation in the optimal points are listed in Table 2.8, where the relative errors of all constrains and objective function demonstrate high accuracy of the performed optimisation.

Table 2.8.

Optimal results							
Symbol, unit		Initial domain			Modified domain		
		Approx.	ANSYS	Δ , %	Approx.	ANSYS	Δ , %
Design parameters	V_{pull} , cm/min		31.0			64.0	
	T_{room} , °C		21.3			40.0	
	P_{mw} , kW		3.42			4.98	
	T_{cont} , °C		150.0			169.3	
Constrains	T_{cent} , °C	185.70	175.80	5.63	165.30	180.90	-8.62
	α_{cent}	0.99	0.97	2.06	0.93	0.98	-5.10
	T_{surf} , °C	163.20	155.10	5.22	166.40	177.00	-5.99
	α_{surf}	0.98	0.94	4.26	1.00	0.95	5.26
	α_{coat} inject	0.75	0.69	8.70	0.75	0.77	-2.60
	T_{coat} , °C	158.40	157.20	0.76	176.20	181.30	-2.81
	α_{coat}	0.93	0.89	4.49	1.00	0.90	11.11
	k_t	0.139	0.141	-1.420	0.153	0.143	6.990
Objective function, Wh/m		203	203	0.00	140	140	0.00

	Pull speed, cm/min	Room and resin temperature, °C	MW Power, kW	Heaters control temperature, °C	Maximal temperature at centerline, °C	Degree of cure at centerline	Maximal temperature at profile surface, °C	Degree of cure at profile surface	Degree of cure on profile surface at coating injection point	Maximal temperature in coating, °C	Degree of cure in coating	Relative time of heaters work k_t	Energy consumption, kWh/m
Optimal	36.9	20.0	3.87	150.6	181.56	0.93	161.77	1.00	0.75	158.10	0.93	0.141	0.191
min	30	20	2	70		0.93		0.93	0.75		0.93		
max	100	20	6	170	190		190		0.85	250			

Fig. 2.30. Optimal results obtained by GRG method for the room temperature of 20 °C.

Based on the results of optimisation, Microsoft Excel tool has been developed for the effective design of microwave assisted pultrusion processes with in-line coating technology. As an example of its application, optimal values of design parameters and corresponding constrains for the room temperature of 20 °C are presented in Fig. 2.30.

2.8. Conclusion

1. The developed and successfully validated numerical approaches for the simulation of pultrusion processes are reliable and can be used for investigation of pultrusion processes. The first procedure has been developed in ANSYS Mechanical environment and is based on the mixed time integration scheme and nodal control volumes method to decouple the coupled energy and species equations. The second procedure has been performed by using ANSYS CFX software. The cure reaction in this case has been introduced as an additional variable. The developed procedures have been successfully validated by the results from literature and physical experiment that gave the possibility to compare the developed algorithms in their applicability for an accurate thermochemical simulation of pultrusion processes without and with temperature control thus defining their advantages and limitations. Good agreement between the present finite element results and published results as well as the results of trial has been observed for the temperature and degree of cure fields in all test problems studying the pultrusion of flat plate, I-beam and two different cylindrical rod profiles. Some difference between experimentally measured and simulated degrees of cure has been observed for the resin at the gelation stage. However, this disagreement practically disappears for the resin which is almost fully cured.
2. The proposed methodology for building of the curing kinetic models has been successfully validated for an epoxy resin described in the available literature. The best precision has been obtained with the Kamal-Sourour model. An engineering tool based on Microsoft Excel code has been developed by using the proposed methodology. This tool has been successfully applied for building of the curing kinetic models of polyester resin and epoxy coating with high microwave absorption properties used in the advanced pultrusion processes.
3. During the experimental validation of the developed algorithms for the simulation of pultrusion processes, it has been shown that the conditions of transient process are very sensitive to the variation of initial data (process parameters). An appropriate methodology for the correction and obtaining of the correct set of initial data using experimental results has been demonstrated.

4. New simulation methodology based on COMSOL Multiphysics and ANSYS Mechanical has been developed for simulation and investigation of advanced microwave assisted pultrusion process. Parameters of the microwave assisted pultrusion process, and the temperature and degree of cure fields in the composite material have been determined for the design of advanced pultrusion die. Simulation results show that the developed technology gives the possibility to ensure a homogeneous curing of the composite profile and to obtain high values of the degree of cure in the cured composite. To improve the effectiveness of the microwave assisted pultrusion process, the influence of different process parameters has been additionally investigated.
5. It has been shown that it is impossible to obtain the complete curing of the profile and applied coating only with the microwave heating taking into account all process conditions and constraints in the case of in-line coating technology. Some electrical heaters should be applied in the pultrusion process to cure coating.
6. It has been shown that process conditions (temperature and degree of cure) are very sensitive to the variation of process parameters (pull speed, applied energy, controlled and room temperatures). Determination of appropriate process conditions is possible only by formulation and solution of optimisation problem. This task has been successfully solved with the purpose to define the minimum of energy consumption. As a result of optimisation, Microsoft Excel tool for determination of optimal process conditions depending on the taken design parameters has been created.

For the future research and design studies, the pultrusion of thin-walled profiles should be studied. Additionally, the thermomechanical problem of the pultrusion taking into account pull forces and friction between the pultruded product and pultrusion die should be considered.

REFERENCES

- [1] Arkhangel'skii I.V., Dunaev A.V., Makarenko I.V., Tikhonov N.A., Belyaev S.S., Tarasov A.V. Non-isothermal kinetic methods. Workbook and laboratory manual. Berlin: Edition Open Access, 2013, 73 p.
- [2] ASTM E 698. Standard test method for Arrhenius kinetic constants for thermally unstable materials.
- [3] Auzins J., Janushevskis A., Janushevskis J., Skukis E. Software EDAOpt for Experimental Design, Analysis and Multi-Objective Robust Optimization. In: *Proceedings of the International Conference on Engineering and Applied Sciences Optimisation*, Kos Island, Greece, 2014, 23 p.
- [4] Auzins, J. Direct Optimization of Experimental Designs. In: *Proceedings of the 10th AIAA/ISSMO Multidisciplinary Analysis and Optimization*, Albany, USA, 2004, V. 274578, 12 p.
- [5] Chachad Y. R., Roux J. A., Vaughan J. G. Three-dimensional characterization of pultruded fiberglass-epoxy composite materials. *Journal of Reinforced Plastics and Composites*, 14, pp. 495–512, 1995.
- [6] Hewlett-Packard: Basics of measuring the dielectric properties of materials, Application Note 1217-1, Hewlett-Packard Co., 1992.
- [7] Hewlett-Packard: HP-Network Analyzer 8720 D + Dielectric Probe Kit 85070B: User's Manual, 1993.
- [8] Howald A.M. Meyer L.S. Shaft for Fishing Rods. US patent US2571717 A, 1951.
- [9] http://www.lucintel.com/pultrusion_market_2021.aspx, 15 August 2017.
- [10] <http://www.synthesites.com/ipage.php?p=9&l=11>, 16 February 2017.
- [11] Janushevskis A., Akinfiyev T., Auzins J., Boyko A. A Comparative Analysis of Global Search Procedures. *Proceedings of the Estonian Academy of Sciences Engineering*, 10 (4), pp. 236–250, 2004.
- [12] Kissinger H. E. Reaction kinetics in differential thermal analysis. *National Bureau of Standards*, 29(11), pp. 1702–1706, 1957.
- [13] Lasdon L. S., Fox R. L., Rather M. W. Nonlinear optimization using the generalized reduced gradient method. *Revue française d'automatique, d'informatique et de recherche opérationnelle. Recherche opérationnelle*, 8(3), pp. 73–103, 1974.
- [14] Lee W.I., Loos A.C., Springer G.S. Heat of reaction, degree of cure, and viscosity of Hercules 3501-6 resin, *Journal of Composite Materials*, 16, pp. 510–520, 1982.
- [15] Liu X. L., Crouch I. G., Lam Y. C. Simulation of heat transfer and cure in pultrusion with a general-purpose finite element package. *Composites Science and Technology*, 60, pp. 857–864, 2000.
- [16] Liu X.-L. Numerical modelling on pultrusion of composite I beam. *Composites: Part A*, 32, pp. 663–681, 2001.
- [17] Myers R. H., Montgomery D. C. Response surface methodology: Process and product optimization using designed experiments. New York: John Wiley & Sons, 1976, 714 p.
- [18] Püschner H. Wärme durch Mikrowellen. Eindhoven: Philips Technische Bibliothek, 1964, 325 p.

- [19] Shokrieh M.M., Aghdami A.M. A dynamic model to simulate the time dependent pultrusion process of Glass/Polyester Composites, *Applied Composite Materials*, 18, pp. 585–601, 2011.
- [20] Sucher M., Fox J. *Handbook of microwave measurements*, Vol. II, 3rd Edition. New York: Polytechnic Press., 1963, 340 p.
- [21] Suranto B. R., Ye L., Mai Y. W. Simulation of temperature and curing profiles in pultruded composite rods. *Composite Science and Technology*, 58, pp. 191–197, 1998.
- [22] Valliappan M., Roux J. A., Vaughan J. G., Arafat E. S. Die and post-die temperature and cure in graphite/epoxy composites. *Composites: Part B*, 27B, pp. 1–9, 1996.
- [23] Witten E. *Composites Market Report 2012: Market Developments, Trends, Challenges and Opportunities*, 2012.
- [24] Wu C.F.J., Hamada M. *Experiments: Planning, Analysis, and Parameter Design Optimisation*. New York: John Wiley & Sons Inc., 2000, 630 p.

3. APPROBATION AND PUBLICATIONS

3.1. List of Publications

1. Barkanov E., Akishin P., Miazza N. L. and Galvez S. (2015): Numerical Simulation of Pultrusion Processes: Algorithms' Comparative Study. – In: Proceedings of the 10th International Conference on Composite Science and Technology (Lisbon, Portugal), 12 p.
2. Akishin P., Barkanov E. and Bondarchuk A. (2015): Finite Element Modelling and Analysis of Conventional Pultrusion Processes. IOP Conf. Series: Materials Science and Engineering, 96, 012012, 10 p (in SCOPUS).
3. Barkanov E., Akishin P., Emmerich R. and Graf M. (2016): Numerical Simulation of Advanced Pultrusion Processes with Microwave Heating. - In: Proceedings of the VII European Congress on Computational Methods in Applied Sciences and Engineering (Eds. M. Papadrakakis, V. Papadopoulos, G. Stefanou, V. Plevris), Vol. IV (Crete, Greece), 7720–7738 (in SCOPUS).
4. Akishin P., Barkanov E., Miazza N. and Galvez S. (2017): Curing Kinetic Models of Resins for Microwave Assisted Pultrusion. Key Engineering Materials, Vol. 721, 92–96 (in SCOPUS).
5. Barkanov E., Akishin P., Miazza N. L. and Galvez S. (2017): ANSYS-Based Algorithms for a Simulation of Pultrusion Processes. Mechanics of Advanced Materials and Structures, Vol. 24, No. 5, 377–384 (in SCOPUS).
6. Barkanov E., Akishin P., Miazza N. L., Galvez S. and Pantelelis N. Experimental Validation of Thermo-Chemical Algorithm for a Simulation of Pultrusion Processes. IOP Conf. Series: Journal of Physics (accepted) (in SCOPUS).

3.2. List of Conferences

1. Akishin P., Barkanov E. and Bondarchuk A. Finite Element Modelling and Analysis of Curing and Pultrusion Processes. International Conference on Physics and Mechanics of New Materials and Their Applications. Azov, Russia, 19–22 May 2015.
2. Barkanov E., Akishin P., Miazza N. L. and Galvez S. Numerical Simulation of Pultrusion Processes: Algorithms' Comparative Study. 10th International Conference on Composite Science and Technology. Lisbon, Portugal, 2–4 September 2015.
3. Akishin P., Barkanov E., Miazza N. and Galvez S. Building of Curing Kinetic Models for a Simulation of Pultrusion Processes. Baltic Polymer Symposium. Sigulda, Latvia, 16–18 September 2015.
4. Akishin P., Barkanov E. and Bondarchuk A. Finite Element Modelling and Analysis of Conventional Pultrusion Processes. 2nd International Conference Innovative Materials, Structures and Technologies. Riga, Latvia, 30 September – 2 October 2015.
5. Barkanov E., Akishin P., Emmerich R. and Graf M. Numerical Simulation of Advanced Pultrusion Processes with Microwave Heating. VII European Congress on Computational Methods in Applied Sciences and Engineering (ECCOMAS 2016). Crete, Greece, 5–10 June 2016.

6. Akishin P., Barkanov E., Miazza N. and Galvez S. Curing Kinetic Models of Resins for Microwave Assisted Pultrusion Processes. 25 International Baltic Conference of Engineering Materials&Tribology (Baltmattrib 2016). Riga, Latvia, 3–4 November 2016.
7. Barkanov E., Akishin P., Miazza N. L., Galvez S. and Pantelelis N. Experimental Validation of Thermo-Chemical Algorithm for a Simulation of Pultrusion Processes. V International Conference on Topical Problems of Continuum Mechanics. Tsaghkadzor, Armenia, 2–7 October, 2017.

3.3. Project Seminars

1. Akishin P., Barkanov E. Simulation of Pultrusion Processes. Under the European Commission 7th Framework Program project COALINE. Brussels, Belgium, 24–25 February, 2014.
2. Akishin P., Barkanov E. Simulation of Pultrusion Processes. Under the European Commission 7th Framework Program project COALINE. Berlin, Germany, 14 May, 2014.
3. Akishin P., Barkanov E. Simulation of Pultrusion Processes. Under the European Commission 7th Framework Program project COALINE. Riga, Latvia, 24–25 September, 2014.
4. Akishin P., Barkanov E. Building of Curing Kinetic Models for a Simulation of Pultrusion Processes. Under the European Commission 7th Framework Program project COALINE. Valencia, Spain, 12–13 February, 2015.
5. Akishin P., Barkanov E. Experimental Validation of Numerical Simulation of Pultrusion Processes. Under the European Commission 7th Framework Program project COALINE. Brussels, Belgium, 30–31 March, 2015.
6. Akishin P., Barkanov E. Design of Multi-Stage and Modular Die. Under the European Commission 7th Framework Program project COALINE. Padova, Italy, 15–16 September, 2015.
7. Akishin P., Barkanov E. Pultrusion of Coated Rod Profile. Under the European Commission 7th Framework Program project COALINE. Bordo, France, 02–03 February, 2016.
8. Akishin P., Barkanov E. Pultrusion of Box Profile. Under the European Commission 7th Framework Program project COALINE. Madrid, Spain, 20–22 September, 2016.
9. Akishin P., Barkanov E. Optimisation of Rod Profile Pultrusion Process. Under the European Commission 7th Framework Program project COALINE. Madrid, Spain, 20–22 September, 2016.

3.4. Approbation of Thesis Results

1. Akishin P. Design of Advanced Pultrusion Processes (Progresīvo pultrūzijas procesu izstrāde). Institute of Mechanics, Riga Technical University, Riga, 25 April, 2017.