Research Article

Open Access

Małgorzata Chwał* and Aleksander Muc*

Simplified optimal modeling of resin injection molding process

<https://doi.org/10.1515/epoly-2019-0039>

Received February 19, 2019; accepted March 08, 2019.

Abstract: A simplified procedure has been developed to optimize the molding processes of thermosetting resins. The proposed methodology involves the classical finite element commercial package (NISA II v. 17). Here, the theoretical background and numerical implementation of the procedure are described. Various design variables that can describe the RIM are discussed. The solved example represents the influence of the heating regime on the curing process. The results are compared with the results obtained with the use of full analysis conducted with the aid of the finite volume analysis (filling and curing stages). They demonstrate the evident advantages of using the simplified optimization approach.

Keywords: RIM process; curing of the thermosetting resin; optimization, finite element analysis

1 Introduction

Currently, manufacturing companies are concentrated on how to further enhance the manufacturing process of a new product or, to speak in terms of time, how to decrease the time-to-market. Therefore, the early connection of a material with appropriate technology in a particular product is essential for it to be considered in further product development steps.

Foundational works by Loos and Springer (1-3), relating to heat transfer and resin cure kinetics, has led to a comprehensive science-based understanding of the thermal response of autoclave and oven cured thermoset materials and/or composites. Although not exhaustive, those results highlight contributions to this area of research that show shifts in our understanding

from science-based knowledge to the use of this science base in practice. Reaction injection molding (RIM) is a processing technique that is commonly applied. RIM combines the polymer synthesis and final shaping into a single operation.

Broyer and Macosko (4) divided the RIM process into mixing, filling and curing stages. The proposed division makes an analysis more convenience. In the whole RIM process, the filling and curing periods form most of the process and are the periods in which changes can be introduced to improve the properties of the final product. Moreover, for complex molds and more reactive mixtures, understanding the filling stage becomes increasingly important (5) and criteria are required to foresee incomplete filling (6).

Highly reactive polymer material processing is currently a fundamental technology to achieve low cycle times and consequently reduce manufacturing costs of parts. However, for complex processes and part geometries, the usually adopted practice considering many design iterations till an acceptable result is obtained can be cumbersome. Therefore, it is desirable to experimentally describe the reaction kinetics to allow model predictions and finally optimize the process by numerical simulation methods. Majority of research works dealing with the optimization of technological processes is devoted mainly to the analysis of curing processes for composites with a thermosetting matrix. A review of existing works is presented, e.g. by Zhang et al. (7). The optimal design of the RIM process is discussed by Muc and Saj (8,9) and Kim et al. (10).

To improve the quality of thermosetting materials which are increasingly used in today's industry, the thermal aspects of the processes have to be optimized. In this article, two various methodologies are proposed to this aim:

- full filling and cure stages analysis based on the use of the finite volume method (FVM),
- cure stage analysis only based on the use of the finite element method (FEM).

The set of governing relations characterizing the filling and cure stages of thermosetting resins is demonstrated in Appendix.

* Corresponding authors: Małgorzata Chwał and Aleksander Muc, Institute of Machine Design, Cracow University of Technology, Warszawska 24, 31-155 Kraków, Poland, e-mail: malgorzata.chwal@pk.edu.pl, olekmuc@mech.pk.edu.pl

2 Description of the heat conduction and the curing process

A comprehensive discussion of the consolidation of thermosetting resins would include many complex phenomena. Simultaneously, there is heat, momentum and mass transfer, accompanied by the chemical curing reaction of the resin (Figure 1).

Using the governing equations presented in the Appendix the heat conduction process of resin materials curing can be written in the following way:

$$\rho_r V_r \frac{\partial}{\partial t}(H) + \lambda_{xx} \frac{\partial^2 T}{\partial x^2} + \lambda_{yy} \frac{\partial^2 T}{\partial y^2} + \lambda_{zz} \frac{\partial^2 T}{\partial z^2} = \frac{\partial}{\partial t}(\rho_r C_p T) \quad (1)$$

where T and t are temperature and time, respectively; λ_{xx} , λ_{yy} , λ_{zz} denote heat transfer coefficients at x , y and z directions; ρ_r , V_r and C_p are the density, volume fraction and the specific heat of the resin; H is the exothermic enthalpy from the resin curing process. The rate of heat generation can be established in the following way:

$$\frac{\partial}{\partial t}(H) = H_r \frac{\partial \alpha}{\partial t} \quad (2)$$

where α is the degree of cure, and H_r is the total amount of heat generated during a complete resin reaction. The following relation expresses the resin curing kinetics:

$$\frac{d\alpha}{dt} = f(\alpha, t) \quad (3)$$

The most often used in practical applications is the model proposed by Kamal and Sourour in 1973 (11). The

model allows for determining the rate of curing reaction depending on the degree of cure α as follows:

$$\frac{d\alpha}{dt} = (k_1 + k_2 \alpha^m)(1-\alpha)^n \quad (4)$$

The reaction rate constants k_1 and k_2 are usually determined from the Arrhenius relationship defined by the formula:

$$k_i = A_i \exp\left(\frac{-E_i}{T}\right) \quad (5)$$

where: $i = 1, 2$; A_i - exponential factors [1/s], E_i - activation energies [K]. The sum of the parameters $m + n$ defines the so-called order of reaction speed.

In Eq. 5 the parameters can be identified from isothermal measurements to separate capacitive and exothermic effects. The typical forms of the degree of cure are presented in Figure 2.

The similar curves were obtained experimentally by Montserrat and Malek (12) from non-isothermal DSC tests. The cited authors mentioned that the form of the curves plotted in Figure 2 was strongly affected by the heating rates.

From the mathematical (numerical) point of view, the RIM analysis can be reduced to the solution of the coupled heat transfer (1) and cure (3) problem. It is evident that numerical modeling (optimization) is more general and applicable to a broader range of problems than analytical solutions. Also, the physical experiments are expensive, prone to errors and time-consuming.

Figure 1: The description of the technological process associated with the curing of resins: A - the gelling threshold, B - curing the area near the infusion channel (blockage of the infusion), C - resin cured in the entire volume.

Figure 2: Distributions of the degree of cure α with physical time t for several heating rates (temperatures to time T/t).

3 Optimal design – objective functions and design variables

The RIM technological processes discussed herein can be schematically presented in the form shown in Figure 3.

The primarily heated resin mixture is introduced into the mold through the injection gate, which number may be larger than one. After that, the mold is heated to the set temperature, consolidated and cooled. The assumed temperature profile is navigated by a finite set of electric heaters distributed around the mold.

In an optimization problem, the optimization process and forms of design variables are mostly coupled with the definition of the temperature regime – see Figure 4.

However, in engineering practice the final quality of the product and residual stresses obtained in the curing process depend on the variety of factors and each of them may be treated as the design variable in the optimization problems. In general, they may be classified in the following manner: (A) resin mixture – type of the resin (polyester, epoxy etc.), types of fillers and hardeners, weight/volume fractions of components, relation between viscosity, time, temperature and degree of cure, (B) form of the mold – geometry, positions, number and type (point or line) of the inlet gates, (C) prescribed parameters of the technological process – velocity of the resin mixture at the input gate, pressure at the input gate, initial temperature of the resin mixture, application of vacuum (or not),

total number of products, control of parameters during the process. They are not independent parameters since everything depends on the existing equipment and the engineering realization of the technological process.

In respect of optimization problems considering the numerical analysis of the Navier-Stokes equations for the technological RIM process two general groups of objectives are present:

- for mold – shape optimization similar to structural shape optimization problems,
- for technological processes – optimization of parameters or boundary/initial conditions.

If the curing process begins around the inlet gate it may result in the shrinkage of the resin in the final stage, and on the other hand, in the lack of the resin in other regions of the mold. Thus, the resin mixture flow may be blocked by the partially cured resin, i.e. the resin having (e.g. according to the commonly accepted assumption) the degree of cure α greater than 65%. To conduct the RIM process in a convenient way, it would be necessary to build the optimal spatial distribution of α parameter. The examples of such curves are drawn in Figure 5 for two points P1 and P2. It is assumed that the point P1 is located in the neighborhood of the inlet gate, and the point P2 far from the inlet gate. If the thermal profile determined by the electric heaters (Figure 3) leads to the curing process in the form presented in Figure 5a. For $t > 400$ sec the cured resin at the point P1 precedes hardening of the

Figure 3: 2-D representation of the RIM process.

resin at the point P2. Changing the thermal profile we can change the sequence of hardening to the form shown in Figure 5b.

Thus, the shaded area in Figure 5a can be treated as the measure of the incorrectness of the curing process. The above graphical representation of the optimization can be formulated in more rigorous mathematical form as follow:

To maximize:

$$F = \sum_{i=1}^{I_h} \frac{\int_0^{t_{Endi}} \alpha_{Pi} dt}{100 \times t_{Endi}} - \sum_{t_i=1}^{N_t} A \times pen \quad (6)$$

where: I_h – the total number of the control points P_i inside the mold (see Figure 3), t_{Endi} – the total time of the curing process at the control point P_i , α_{Pi} – the curing degree at the control point P_i , t_i – the i -th time step, N_t – the total number of time steps, pen – the penalty coefficient, A – the value computed from the formula:

$$A = \begin{cases} \frac{d\alpha}{ds_p} & \text{if } \frac{d\alpha}{ds_p} > 0 \\ 0 & \text{if } \frac{d\alpha}{ds_p} < 0 \end{cases} \quad (7)$$

Let us note that the integrals in Eq. 6 are normalized to the value $100 \times t_{Endi}$ representing the hypothetical rectangle (the infinite speed of curing) to eliminate the situations where the curing goes very slowly. The second term A is understood as the measure of correctness of curing direction. In this way, the objective function should fulfil the required sequence of the curing processes at the control points with the simultaneous reasonable speed of the RIM process at the control points P_i .

The general overview of the optimization analysis is shown in Figure 6. In general to evaluate the objective function the finite element volume or method can be used. However, the FEV method is much more general since it allows us to describe accurately two stages of the RIM process, i.e. filling of the mold and curing the resin – see Figure 1. Using the FEM, it is possible to characterize the second stage of the process, i.e. consolidation of the resin only. The effectiveness of searching for optimal solutions is defined by the form of design variables and optimization algorithms. Various types of optimization algorithms are characterized by Muc (13). In the present paper, the optimization problems are solved with the use of genetic algorithms (14) or evolutionary algorithms (15).

Using the FVM (e.g. Fluent) at each time step one can evaluate the components of the fluid velocity vector, viscosity, degree of cure and temperature for each of finite volumes. Although many general-purpose FE packages have a facility to perform transient heat transfer analysis

Figure 4: Temperature vs time distribution during the manufacturing process.

a) the incorrect curing process

b) the optimal curing process

Figure 5: Distributions of the degree of cure α .

Figure 6: The graphical representation of the optimization problem.

(1), not all of them have the facilities for resin-cure simulation (3). Therefore, as it reported, e.g. by Joshi et al. (16), the degree of cure (3) is computed approximately (the finite difference approximation) at each step of time, and then the heat transient problem (1) is solved. At each time step, the distributions of the temperatures and the degree of cure are determined. However, similarly as in the FVM the success of the iterative FEM procedures depends on the number of iterations and the length of time steps involved. Such a situation is not convenient from the optimization point of view. Therefore instead of searching for the optimum of the objective function (6), we propose to simplify the optimization problem by its reduction to the FE heat conduction analysis without calculation of the degree of cure – a geometrically linear analysis.

The spatial degree of cure distributions are controlled by the temperature distributions and the heating rates – see Figure 2. It seems to be reasonable to model the temperature distributions optimally, i.e. satisfying the criterion (6). It may be achieved by solving the following problem:

To find the global optimum:

$$\text{Max } T(x,y,z) \quad (8)$$

in the region S the most far removed from the inlet gate.

It is assumed that the allowable solutions do not have the local minima/maxima in the design space. Let us note that the optimization problem (8) can be solved using the classical packages (ANSYS, ABAQUS, NISA II). Then, the correctness of the optimal design can be verified with the FVM.

4 Numerical example

To illustrate the effectiveness of the proposed simplified optimization methodology the planar problem analogous to that discussed by Muc and Saj (8,9) has been solved. The geometry of the mold and the manufactured part is shown in Figure 7.

In the optimization problem, the design variables are defined by the temperature at six electric heaters (H1-H6). For the RIM processes, commonly the temperature is constant. For the temperatures equal to 140°C the distribution of degree of cure is demonstrated in Figure 8. The sequence of the degree of cure variations with time for 5 selected points (P1-P5) is presented in Figure 9.

The maximal temperature occurs at the middle of the manufactured parts, and it corresponds to the quickest consolidation of the resin (the points P3 and P4), although the resin hardening at the point P5 (close to the inlet gate) develops rather slowly. In this case, the value of the objective function is equal to 2.98.

It is assumed that the variations of temperature are constrained (technological requirements of the RIM process) and lie in the interval $[T_{min}, T_{max}]$, where $T_{min}=100^{\circ}\text{C}$, and $T_{max}=170^{\circ}\text{C}$. The detailed optimization results conducted with the use of the full, FV method are summarized in Table 1 and compared with the optimal values of design variables derived with the aid of the simplified analysis and the objective function in the form (8).

The comparison of the results demonstrates a very good agreement between full and simplified methods of optimization. The real advantageous benefit of the proposed method depends mainly on the significant

Figure 7: The geometry of the analyzed problem.

Figure 9: Variations of the degree of cure for the controlling points – the constant temperature at the electric heaters (140°C).

Figure 8: Distribution of degree of cure for the constant temperature at the electric heaters (140°C).

reduction of the computational time and the simplicity of the formulation and solving of the optimization problem. In the simplified method, the classical FE packages

can be used with the implementation of the classical simplest gradient method of optimization. The problems considered have been solved with the aid of the NISA II (version 17) FE package.

For the comparison of the results, the distribution and variations of degree of cure for the optimal temperature are plotted in Figures 10 and 11.

If the curing process is optimized the control point P5 has the lowest degree of cure (see Figures 10 and 11, respectively) and it allows us to fill the mold by the resin almost to the end of the process.

5 Conclusions

The simplified optimal selection strategy of searching for a non-uniform, spatial temperature distribution around the mold (but constant in time) to facilitate the resin flow during the mold filling process of RIM was proposed. Then, the results were compared with complete computations including both filling and curing stages. The results demonstrated the evident advantages of the proposed simplified design based mainly on: the simplicity of the computational finite element analysis (the use of finite volume method is not required) and the drastic reduction of the computational time.

Table 1: Comparison of optimal designs conducted for full and simplified optimization methods.

Number of the electric heater	Full (FVM) optimal design temperature T [°C]	Simplified (FEM) optimal design temperature T [°C]
H1	100.1	100.0
H2	170.0	170.0
H3	170.0	170.0
H4	166.9	167.5
H5	156.8	155.0
H6	165.6	165.0
Number of iterations	32	12
Value of the objective function (6)	4.172	4.176
Computational time	54.6 min	84 sec

Figure 10: Distribution of degree of cure for the optimal temperature at the electric heaters (Table 1).

The proposed procedure can be extended with easy to other class of optimization problems related to the process variables in the RIM technology – see the paper by Muc et al. (17). We also intend to implement a similar methodology in the optimization of the composite production technologies, e.g. the resin transfer molding or autoclave methods – see the papers by Muc and Chwał (18,19).

Appendix: The governing physical relations characterizing the RIM process

Considering the non-isothermal process, the fundamental equations should characterize at the same time different phases of the technological process, namely: (1) the behaviour of the fluid (resin) during the filling of the mold together with (2) a heat transfer analysis from the mold walls and (3) the reactive nature and rheology of reactive materials (the viscosity and curing kinetics) during the gelation stage of the process. The Navier-Stokes equations are usually used in the modeling of such class of problems. However, a set of additional relations describing the viscosity and curing kinetics is added to the Navier-Stokes equations. The fundamental Navier-Stokes equations for the viscous Newtonian fluid may be written in the integral form as:

$$\frac{\partial}{\partial t} \int_{\Omega} \vec{W} d\Omega + \oint_{\partial\Omega} (\vec{F}_c - \vec{F}_v) dS = \int_{\Omega} \vec{Q} d\Omega \quad (A1)$$

alternatively, in the equivalent differential form:

$$\frac{\partial \vec{W}}{\partial t} + \nabla \cdot (\vec{F}_c - \vec{F}_v) = \vec{Q} \quad (A2)$$

where:

$$\begin{aligned} \vec{W} &= [\rho, \rho V_j, \rho E], \quad \vec{F}_c = [\rho \vec{V}, \rho V_i V_j + p \vec{n}, \rho H \vec{V}], \\ h &= cT, \quad \vec{F}_v = [0, \tau_{ij}, \Theta_i], \\ \vec{Q} &= [0, \rho f_i, \rho f_i V_i + \dot{q}_h], \quad \Theta_i = V_j \tau_{ij} + \lambda \frac{\partial T}{\partial x_i}, \\ H &= h + \frac{1}{2} V_i V_i = E + \frac{p}{\rho} \end{aligned} \quad (A3)$$

Figure 11: Variations of the degree of cure for the controlling points – the optimal temperatures at the electric heaters (Table 1).

These relations describe the exchange (flux) of mass, momentum and energy through the boundary $\partial\Omega$ of fixed in space a control volume Ω . There are two flux vectors $\vec{F}_c$, $\vec{F}_v$. The vector $\vec{F}_c$ is related to the convective transport of the quantities in the fluid. Usually, it is termed vector of convective fluxes, while for the momentum and the energy equation it also includes the pressure terms $p\vec{n}$ where p denotes a static pressure and $\vec{n}$ is a unit, normal vector. $\vec{V}$ denotes the velocity vector having the components V_j . The vector $\vec{F}_v$ is the vector of viscous fluxes, contains the viscous stresses τ_{ij} defined as follows:

$$\tau_{ij} = \mu \left(\frac{\partial V_i}{\partial x_j} + \frac{\partial V_j}{\partial x_i} \right) - \frac{2}{3} \mu \frac{\partial V_l}{\partial x_l} \delta_{ij} \quad (A4)$$

where T – temperature, λ – thermal conductivity coefficient, μ – molecular fluid viscosity coefficient, H – the total enthalpy, h – the enthalpy, f_i – components of the body forces, $\dot{q}_h$ – the time rate of the heat transfer per unit mass, E – the total energy of a fluid per unit mass.

The presented general relations, which describe a resin flow and a heat transfer are supplemented by the equations describing the rheokinetics of the resin (the viscosity relations):

$$\eta = \frac{\mu}{\rho} = B \exp \left(\frac{T_b}{T} \right) \cdot \left(\frac{\alpha_g}{\alpha_g - \alpha} \right)^{C_1 + C_2 \alpha} \quad (A5)$$

where B – viscosity, C_1 and C_2 – constants, $T_b = E/R$, R – gas constants.

References

1. Lee W.I., Loos A.C., Springer G.S., Heat of reaction, degree of cure, and viscosity of Hercules 3501-6 resin. *J. Compos. Mater.*, 1982, 16, 510-520.
2. Loos A.C., Springer G.S., Curing of epoxy matrix composites. *J. Compos. Mater.*, 1983, 17, 135-169.
3. Loos A.C., Springer G.S., Calculation of cure process variables during cure of graphite/epoxy composites. In: C. Browning (Ed.), *Composite Materials: Quality Assurance and Processing*, no. STP28542S. ASTM International, West Conshohocken, PA, USA, 1983, 110-118.
4. Broyer E., Macosko C.W., Heat transfer and curing in polymer reaction molding. *AIChE J.*, 1976, 22, 268-276.
5. Castro J.M., Macosko C.W., Tackett L.P., Steinle E.C., Critchfield F.E., Premature gelling in RIM. *Soc. Plast. Eng. Tech. Papers*, 1980, 26, 422-427.
6. Macosko C.W., Fundamental concepts in reaction injection molding. In: *Proceedings of the First International Congress on Reactivity of Polymer Processing*. Pittsburgh, USA, October 1980.
7. Zhang J., Xu Y.C., Huang P., Effects of cure cycle on curing process and hardness for epoxy resin. *Express Polym. Lett.*, 2009, 3, 534-541.
8. Muc A., Saj P., Optimization of the reactive injection molding process. *Structural Multidiscipl. Optimiz.*, 2004, 27, 110-119.
9. Muc A., Saj P., Genetic algorithms in optimization of resin hardening technological processes. In: Burczyński T., Osyczka A. (Eds), *Proceedings of IUTAM Symposium on Evolutionary Methods in Mechanics*. Krakow, Poland, 2004, 287-296.
10. Kim H.G., Son H.J., Lee D.K., Kim D.W., Park H.J., Cho D.H., Optimization and analysis of reaction injection molding of polydicyclopentadiene using response surface methodology. *Korean J. Chem. Eng.*, 2017, 34, 2099-2109.
11. Kamal M.R., Sourour S., Kinetics and thermal characterization of thermoset cure. *J. Polymer Eng. Sci.*, 1973, 13, 105-114.
12. Montserrat S., Malek J., A kinetic analysis of the curing reaction of an epoxy resin. *Thermochim. Acta*, 1993, 228, 47-60.
13. Muc A., Evolutionary design of engineering constructions. *Latin American J. Sol. Struct.*, 2018, 15, 1-21.
14. Muc A., Gurba W., Genetic algorithms and finite element analysis in optimization of composite structures. *Compos. Struct.*, 2001, 54, 275-281.
15. Muc A., Muc-Wierzoń M., An evolution strategy in structural optimization problems for plates and shells. *Compos. Struct.*, 2012, 94, 1461-1470.
16. Joshi S.C., Liu X.L., Lam Y.C., A numerical approach to the modeling of polymer curing in fibre-reinforced composites. *Compos. Sci. Technol.*, 1999, 59, 1003-1013.
17. Muc A., Romanowicz P., Chwał M., Description of resin curing process – formulation and optimization. *Polymers*, 2019, 11, 127.
18. Muc A., Chwał M., Analytical discrete stacking sequence optimization of rectangular composite plates subjected to buckling and FPF constraints. *J. Theor. Appl. Mech.*, 2016, 54, 423-436.
19. Chwał M., Muc A., Transversely isotropic properties of carbon nanotube/polymer composites. *Compos. Part B-Eng.*, 2016, 88, 295-300.