

Material Concepts and Processing Methods for Electrical Insulation

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Abstract—Epoxy composites are broadly used as an electrical insulation for the high voltage applications since only such materials can fulfill particular mechanical, thermal, and dielectric requirements. However, properties of the final product are strongly dependent on proper manufacturing process with minimized material failures, as too large shrinkage, voids and cracks. Therefore, application of proper materials (epoxy, hardener, and filler) and process parameters (mold temperature, filling time, filling velocity, initial temperature of internal parts, gelation time), as well as design and geometric parameters are essential features for final quality of the produced components. In this paper, an approach for three-dimensional modeling of all molding stages, namely filling, curing and post-curing is presented. The reactive molding simulation tool is based on a commercial CFD package, and include dedicated models describing viscosity and reaction kinetics that have been successfully implemented to simulate the reactive nature of the system with exothermic effect. Also a dedicated simulation procedure for stress and shrinkage calculations, as well as simulation results are presented in the paper. Second part of the paper is dedicated to recent developments on formulations of functional composites for electrical insulation applications, focusing on thermally conductive materials. Concepts based on filler modifications for epoxy electrical composites have been presented, including the results of the obtained properties. Finally, having in mind tough environmental regulations, in addition to current process and design aspects, an approach for product re-design has been presented focusing on replacement of epoxy material with the thermoplastic one. Such “design-for-recycling” method is one of new directions associated with development of new material and processing concepts of electrical products and brings a lot of additional research challenges. For that, one of the successful products has been presented to illustrate the presented methodology.

Keywords—Curing, epoxy insulation, numerical simulations, recycling.

I. INTRODUCTION

THERMOSETTING materials are dominant ones in manufacturing of electrical products. In most of the formulations a filler in form of silica or alumina powder is used. This results in lower thermal expansion and reduced shrinkage of such composite, its better thermal conductivity, improved mechanical properties, and also lower cost. Easy processability is another convincing argument for the application of these materials. So called Automated Pressure Gelation (APG) process is mostly used in the manufacturing of electrical products, although for larger components vacuum casting is preferable. In the APG (Fig. 1), two or more liquid

reactants (epoxy resin, hardener) with additional components (filler, accelerator, plasticizer) are mixed together. After homogenizing and degassing, the mixture is injected into the heated mould. Polymerization of the resinous material generates additional heat due to exothermic reaction and the component becomes solidified (cured) obtaining a desired shape. Afterwards, de-molding is done and the secondary heat treatment, aiming curing completion is carried out - very often in a tunnel furnace.

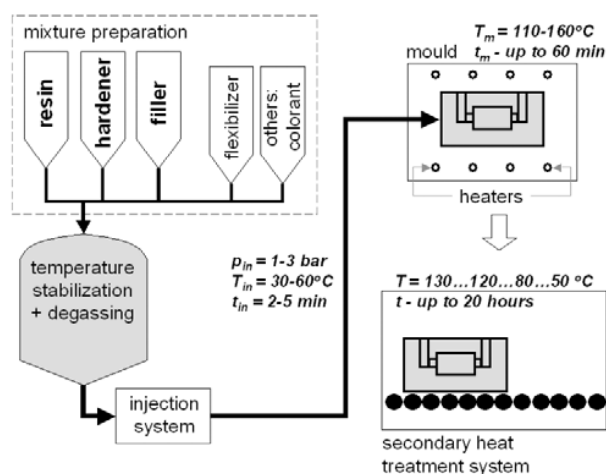


Fig. 1 Principles of APG process

Application of appropriate materials, process parameters, as well as design and geometric parameters are key factors for better quality components. Predicting the most suitable curing conditions for a defined composition of the resinous mixture is possible only on the basis of the examination of changes in its properties, during the course of reactions proceeding at different temperatures and with the optimum parameters fixed. Thus, the knowledge of the parameters effecting the course of curing of thermoset materials is, from the process point of view, of vital importance, permitting the optimization of the process parameters of the production of thermoset components.

Information about degree of curing and the course of the curing phenomenon determines the quality of epoxy resin products and allows to avoid unfavorable directions of curing propagation which encourages the formation of cracks and the weakening of the dielectric properties of products made of these resins. This method which is very useful for selection of the proper process parameters of such reactive molding technology is based on the computer simulation of the process.

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In most cases, computer simulations of the filling and curing stages in the APG technology were based on 2D and 2.5D simulations and it is well established mainly for injection molding technology. However, to have a full picture of the process, such approach is not sufficient and has never been successfully applied in industrial scale for bulk epoxy products, where exothermic effect of the ongoing reaction is very strong. Thus, a development of fully three-dimensional approach was necessary for reactive molding equipment.

II. SIMULATION PROCEDURE

The fully three-dimensional CFD simulation approach relies on modeling of the non-isothermal two-phase fluid flow, that is based on well-known from the literature set of differential equations: continuity, momentum and energy. However, having in mind the reactive nature and complex rheology of reactive materials there is a need for appropriate

characterization of the viscosity and curing kinetics. For that purpose, two empirical models have been selected, and they have been implemented in the CFD code, ANSYS FLUENT, as so called user-defined-subroutine.

FLUENT calculations have been performed until degree of curing exceeds gelation point (α_g). After that stage of modeling, structural analyses have been done and they required an application of another software, dedicated to such calculations. For that, ABAQUS code was selected. The results obtained in CFD calculations (temperature and degree of curing) had to be transferred into ABAQUS as initial conditions (see Fig. 2). Since no adequate direct data transfer codes were available on the market, external data transfer procedure has been developed and successfully implemented to perform the solution mapping between FLUENT and ABAQUS models.

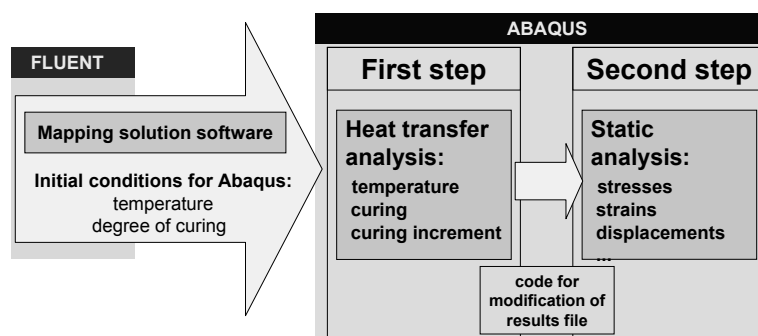


Fig. 2 Sequential simulation approach in reactive molding

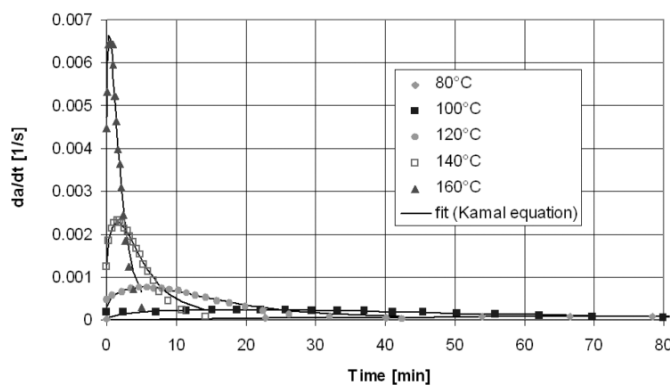


Fig. 3 Degree of cure rates during isothermal cure

A. Curing Kinetics and Viscosity

To determine kinetics of the curing process, Kamal's model is often used [1] and it has been selected in the simulations. According to this model, a degree of curing at time t is defined as:

$$\alpha = H(t) / H_{\Sigma} \quad (1)$$

where; $H(t)$ is heat of reaction released at time t , H_{Σ} is total

heat of reaction.

The equation describing curing rate is the following:

$$d\alpha / dt = (k_1 + k_2 \alpha^m)(1 - \alpha)^n \quad (2)$$

$$k_i = A_i \exp(E_i / RT) \quad (3)$$

where; $i = 1, 2$; $d\alpha/dt$ is curing rate, m, n is constants, k_i is reaction rate constants, A_i is preexponential factors, E_i is

activation energies, R is the universal gas constant, T is the absolute temperature

The sample results of curing kinetics obtained during DSC (Differential Scanning Calorimetry) measurements are presented in Fig. 3.

The rheology of reactive materials requires appropriate characterization of the viscosity and curing kinetics. Properties of material depend on its temperature and degree of curing α . Therefore, to properly simulate the rheology of the epoxy materials, one must use models based on the fact that material viscosity depends not only on temperature, but also on degree of curing. This relation was described by Macosko [2]:

$$\eta = B \exp(T_b / T) ((\alpha_g / (\alpha_g - \alpha))^{C_1 + C_2 \alpha}) \quad (4)$$

where; B , C_1 , C_2 , T_b are constants, $\alpha_g = 0.55-0.80$ (gelation threshold).

The rheokinetics parameters B , C_1 , C_2 and T_b were obtained

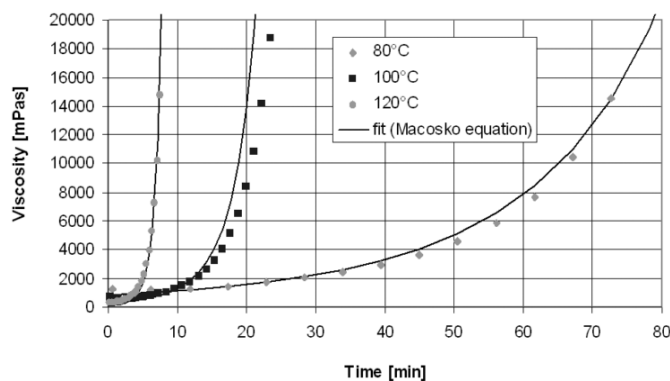


Fig. 4 Viscosity changes during isothermal cure

The major effort was put in taking chemical shrinkage into account. In general, total strain increment (in each time step) can be expressed as a sum of mechanical and thermal components:

$$\Delta \varepsilon_{xx}^{Total} = \Delta \varepsilon_{xx}^{Mech} + \Delta \varepsilon_{xx}^{th} \quad (5)$$

Abaqus software allows defining a thermal component by applying user-defined subroutine (UEXPAN). This component was set to cover chemical as well as thermal effects, which influence a material density:

$$\Delta \varepsilon_{xx}^{th} = \sqrt[3]{\rho' / \rho} - 1 \quad (6)$$

where ρ' means actual density and ρ is a density from previous step. To utilize this equation, it was necessary to know dependency of material density on temperature and curing. This dependency was derived based on experimental measurements. More information regarding the simulation methodology can be found in [3], [4].

by measuring the cure behavior using a dynamic mechanical spectrometer (DMS), Fig. 4.

The results presented refer to the material based on epoxy derived from bisphenol A (DGEBA) CY 228 hardened with HY918 (derived by Huntsman) was used. Of course, the proposed simulation procedure can be used to any thermoset formulation since in the implemented models material parameters can be defined by the user.

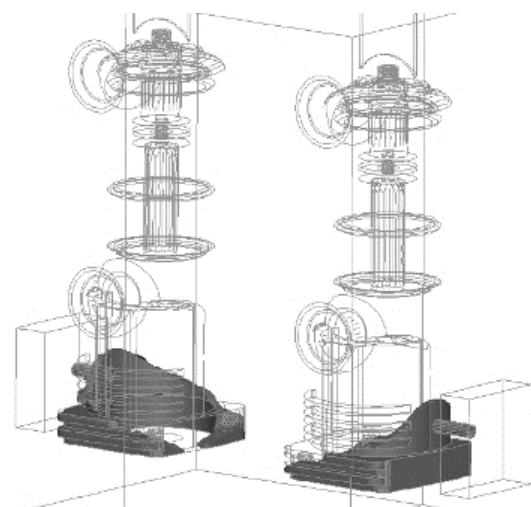
B. Structural Analyses

Structural calculations are performed in sequentially coupled manner. In the first step, only a heat transfer analysis is done (including reaction kinetics) and then in the second step, stress analysis (based on results obtained in the first step) is completed.

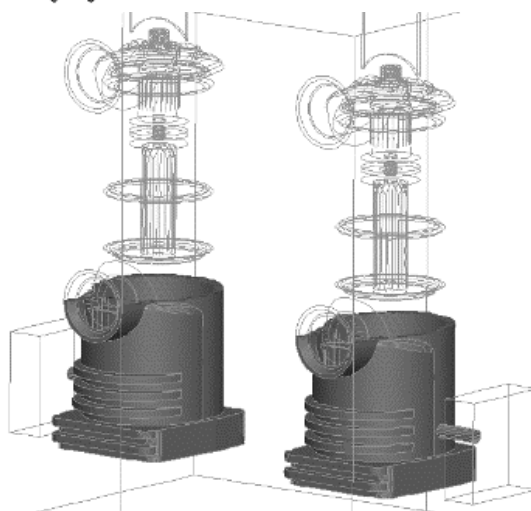
III. SIMULATION RESULTS

Using a proposed simulation approach number of valuable results can be obtained and used for set up the best process parameters. In Fig. 5, sample simulation results illustrating flow pattern have been presented. However, the most interesting information is obtained from the curing results. It is possible to visualize the curing front propagation, and in Fig. 6 such information is presented. The most desired curing behavior is characterized by only one main front moving from the top of the component and the area of epoxy injection should be solidified at the end. A detailed analysis of the obtained results lead to better quality of the cast components since any potential problems can be identified early enough before the mold is ordered and manufactured. Very often the simulation results can be used to reduce cycle time of the manufacturing process by selection of the most suitable mold heating conditions. Also, the simulation of mechanical behavior can bring an important information, especially for product designers. When using numerical simulations, mechanical hot spots (stresses, deformations) can be identified virtually, and before mold fabrications and real manufacturing process, but potential problems can be avoided, and necessary

design changes can be done. Example of stress distribution in the embedded pole is presented in Fig. 7.



Contours of Volume fraction (resin) (Time=1.1250e+01)
Filling stage: volume fraction of resin



Contours of Volume fraction (resin) (Time=3.6650e+01)
Filling stage: volume fraction of resin

Fig. 5 Mold filling profiles

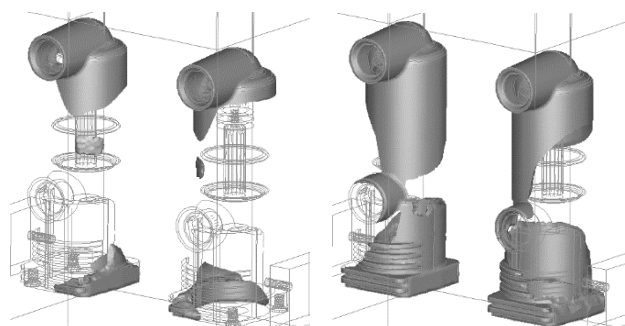
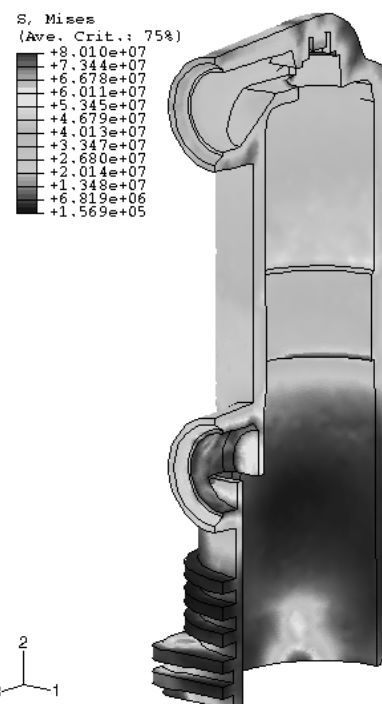
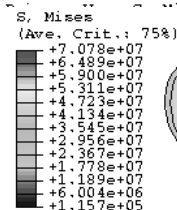


Fig. 6 Curing propagation profiles



Step: Cooling
Increment 21: Step Time = 6650.



Step: Cooling
Increment 28: Step Time = 5.2420E+04

Fig. 7 Generation of internal stresses in epoxy component

IV. MATERIAL MODIFICATION

Pure epoxy resins are characterized by a very low thermal conductivity (ca. 0.2 W/mK) and their direct application in electrical insulation is limited due to poor cooling capabilities. The addition of filler particles into epoxy resin can result not

only in a better mechanical behavior but also in a significant improvement in the thermal conductivity. Generally, thermal properties of the composite are related to size and content of the filler grains and also to thermal conductivity of matrix and the incorporated filler. However, there are very important characteristics of the filler-epoxy matrix interface due to strong phonon scattering processes and occurring interface resistance. To increase thermal conductivity in epoxy composite materials, the conductive paths must be maximized and thermal contact resistance decreased. To obtain high thermal conductivity of the epoxy composites, one can add the fillers that have a very high thermal conductivity (e.g. boron nitride or silicon nitride). However, due to high cost of such materials that solution is not economically accepted.

One of the promising approaches to obtain composite with higher effective thermal conductivity is the modification of filler particles. In contrast to traditional polymer composites in which single filler is usually used, novel core-shell filler material has been proposed in which particles consist of:

- core - made of standard filler material like SiO_2 or Al_2O_3
- shell - obtained by coating with thin layer of a high-thermal-conductivity (HTC) material, mainly boron nitride and aluminum nitride.

Filler materials have been synthesized by means of carbothermal reduction and nitridation process. A representative set of epoxy matrix samples filled with core-shell material has been prepared and compared with the reference samples. The thermal conductivity measurements have been performed on these samples at the room temperature, and sample results are presented in Table I.

TABLE I
THERMAL CONDUCTIVITY OF EPOXY COMPOSITES

Sample	Filler content, vol. %	λ , W/mK	λ increase, %
SiO_2	31	0.62	-
$\text{SiO}_2@\text{Si}_3\text{N}_4$	31	0.92	48
Al_2O_3	31	0.75	-
$\text{Al}_2\text{O}_3@\text{AlN}$	31	1.17	56
Al_2O_3	45	1.40	-
$\text{Al}_2\text{O}_3@\text{AlN}$	45	2.28	63

As it can be noticed, the effective thermal conductivity of composites filled with silica covered with Si_3N_4 as HTC shell increases significantly by 48% in comparison with pure SiO_2 . Composites based on aluminum oxide with AlN as the shell show even a larger enhancement of thermal conductivity. The sample with 31% filler content by volume showed an increase by 56% and the sample filled 45% by volume - by 63% respectively [5].

V. NEW DESIGN AND MANUFACTURING

Having in mind environmental regulations, recently a new trend in design of electrical apparatus has been observed, namely replacement of thermosetting insulation with thermoplastic one, and ABB is very active in that area contributing to reduction the impact of ABB's products on the environment and satisfying the customers' requirements to

fulfill tougher environmental regulations. When using thermosets, during curing, it undergoes a chemical reaction that results in infusible and insoluble network. In case of thermoplastic insulation, it is not a problem since the material can be melted. This opens totally new opportunities for recycling of such electrical components with thermoplastic housing. However, it must be underlined that the whole process related with material change is not so straightforward procedure and requires a comprehensive re-design procedures including manufacturability aspects. Such design for the recycling methodology has been developed and implemented for various electrical products, and in the paper an example of medium-voltage embedded pole has been presented.

To make a replacement from epoxy to thermoplastic, the outer dimensions of the pole were kept the same as for epoxy material. The high pressure injection molding process was used and process parameters were optimized using 3D simulations to obtain good quality product. Thermoplastic material with short glass fibers was used to fulfil mechanical requirements of the pole. Application of the thermoplastic material brought to weight reduction of the complete pole by 35% compared to currently used solution. In Fig. 6, profiles of mold filling during injection molding have been presented. Number of design variants and injection molding arrangements was tested.

Another advantage of the injection molding process was possibility of process automation using robotized station.



Fig. 8 Mold filling patterns during injection molding

In Fig. 9, comparison of epoxy embedded pole and thermoplastic one is presented. More information about that development can be found in [6].



Fig. 9 Embedded pole case – from epoxy to thermoplastic

VI. CONCLUSION

Application of advanced numerical simulations for

modelling of reactive molding process can bring a lot of useful information for designers and technologists dealing with manufacturing of electrical apparatus. Such virtual prototyping can be useful both, in design optimization and in selection of best process parameters during real manufacturing operations.

Another important aspect is related with cooling challenges of electrical devices based on epoxy insulation. Due to a very low thermal conductivity of the insulation, novel concepts based on modification of filler properties can open new opportunities in area of efficient heat dissipation and can avoid overheating problems.

To bring more environmentally friendly solutions for electrical insulation, one of the most promising solutions is replacement of thermosets with thermoplastics. Such approach can offer much faster manufacturing process, better repeatability in the material properties, as well as weight reduction of the products. And also much easier recycling opportunities.

REFERENCES

- [1] M.R. Kamal, "Kinetics and thermal characterization of thermoset", *Polymer Engineering and Science*, 1973.
- [2] C.W. Macosko, *Fundamentals of Reaction Injection Molding*, Hanser Verlag, 1989.
- [3] R.Sekula, P. Saj, et.al, 3-D modelling of reactive molding process: from tool development to industrial application", *Advances in Polymer Technology*, Vol.22, No. 1, 2003.
- [4] R. Sekula, T. Nowak, O. Claus, "Sequential fluid dynamics and structural mechanics simulations of reactive molding process", *International Journal of Materials and Product Technology*, Vol.40, No. 3-4, 2011.
- [5] K. Gaska, "Enhanced thermal conductivity of epoxy-matrix composites", PhD dissertation, University of Science and Technology, Krakow, 2014.
- [6] T. Fugel, D. Gentsch, A. Klaska, Ch. Meyer, "Breaking ahead of expectations", *ABB Review*, No. 1, 2010.