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Experimental Analysis and Inverse Approach in Numerical Modelling of Curing Process of Composite Materials

Ph.D. Thesis

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Abstract

Curing reaction is an inseparable phenomenon connected with the processing of thermosetting materials like for example epoxy resins. These materials constitute an excellent electrical barrier and, additionally, represent very good mechanical and thermal properties. This is the main reason why epoxy resins are widely used in energy industry, where strict requirements are set for power products regarding their reliability and quality, since this affects directly the operation of the whole power transmission and distribution system. One can find electrical bushings, current and voltage transformers, sensors, cable joints, etc. among the products, where epoxy resins are used as electrical insulation.

Such wide application of these thermosets means that the material properties must be tailored to the specific product. For this reason inorganic fillers like silica are used to modify the material properties. Addition of filler can result in higher hardness of the mixture, in lower shrinkage during the mixture curing, in decreased heat generation during the exothermic curing reaction, in the material transparency or its specific colour or, finally, in the improvement of other parameters influencing the material processing and its further exploitation as well as in lower cost of the material. Unfortunately, utilization of fillers is connected with the risk of sedimentation phenomenon that, consequently, can lead to anisotropic properties of electrical insulation what is highly undesirable.

This problem concerns mainly the power products having big dimensions like high-voltage electrical bushings reaching in some cases even more than ten meters of height. This is one of the reasons why the standard epoxy resin insulation has been replaced in the mentioned high-voltage bushings with the composite material in the form of crepe paper impregnated with epoxy resin. Consequently, the manufacturing of electrical insulation, including course of the curing reaction, has changed because of the difference between the material properties of standard epoxy resin and paper-resin composite. Meanwhile, the right execution of the cross-linking process is one of the main aspects influencing the final properties of electrical insulation, mainly because of strong exothermic character of the cross-linking reaction. For example, too high heat generation during the production of epoxy resin insulation can lead to the material

overheating and, simultaneously, to the degradation of the material insulation properties.

The complexity of the curing reaction makes its experimental analysis highly difficult, even in case of epoxy resin mixtures with relatively simple structure. Usually, Differential Scanning Calorimetry (DSC) is used to determine the kinetics of the cross-linking reaction, however, this measurement method is limited by a small size of the investigated sample (few milligrams). Hence, it is doubtful if such little sample is representative for much bigger systems having composite structure like in case of the mentioned paper impregnated with epoxy resin present in high-voltage bushings.

In the connection to above it was decided to make an attempt within the presented thesis to work out an alternative method of determination of the curing reaction kinetics of complex and big composite structures. For this purpose numerical modelling and inverse methodology supported with experimental measurements was proposed. It was possible, since the computational time is not the limiting factor anymore due to, observed already in the 80's, dynamic development of the mentioned inverse methods, of commercial software dedicated to numerical simulations and of processor capacity offered by modern.

The scope of this Ph. D. thesis includes among others the literature review focused on the application of inverse methods, utilizing numerical modelling and experiments, for determination of the kinetics of the phase change process, similar to the curing reaction. Information about electrical bushings and electrical insulation applied in these devices is also a part of introduction. The theoretical basics of inverse analysis and the mathematical description of the curing reaction kinetics can be found as well. Finally, the mentioned DSC, as one of the most famous measurement methods of the curing kinetics determination, is also described.

The preparation of the experimental stand and the execution of the curing experiments for different research samples was the substantial element of this thesis. Both systems with relatively simple and uniform structures like in case of standard epoxy resin as well as complex composite specimens in the form of the mentioned resin impregnated paper were investigated. The goal of this experimental part was twofold, i.e. to qualify how the structure of the investigated samples influences course of the cross-linking reaction and to capture data needed in the subsequent stages of the thesis.

The built up of the mathematical and numerical models describing the performed curing experiments was one of them. In particular, the model of the curing reaction kinetics had to be developed at this stage. For this purpose the inverse analysis was proposed. Basically, in this approach the temperatures recorded during the course of the curing process were applied to determine the parameters of the mentioned model. At this stage the influence of the exothermic curing reaction (resulting in the significant heat generation) on the measured temperature field was important. Additionally, application software was developed within the worked out inverse approach to fully automate the optimization procedure (including computer simulations) executed in each inverse analysis. The application was based on two optimization algorithms, namely Levenberg-Marquardt method and Particles Swarm Optimization.

The developed approach to curing modelling, utilizing the inverse methodology to find the curing kinetics data, was subjected to credibility analysis. In the first step the preliminary tests were conducted to work out the optimum configuration of the optimization algorithms. For this purpose the coefficients estimation problem was solved for two benchmark functions. The goal of the next study, referred to as the virtual curing experiment, was to calculate the unknown curing kinetics parameters within inverse analysis. At this stage the needed measured temperatures were generated by using the mentioned numerical model of the curing process and the known curing kinetics data (so-called simulated measurement). The found curing kinetics model improved the correlation of results with the measurement data confirming the correctness of the principle of working of the proposed inverse methodology.

The next step constituted the experimental validation and this time the temperatures measured in the curing experiment were utilized as additional information required in inverse analysis to determine the parameters of the kinetics model of the cross-linking reaction. It should be noticed that two different kinetics models were worked out at this stage, namely the one describing the course of the curing reaction of the investigated epoxy resin and the second one dedicated to the composite structure consisted of the crepe paper impregnated with the mentioned epoxy resin. In both cases the applied inverse methodology led to the curing kinetics data giving an improved agreement between the simulation and experimental results. Additionally, it was concluded that the model describing the curing kinetics of the epoxy resin cannot be directly applied to the modelling of the cross-linking process of the resin-paper structures.