PREDICTING THE FLAMMABILITY OF EPOXY RESINS FROM THEIR STRUCTURE AND SMALL-SCALE TEST RESULTS USING AN ARTIFICIAL NEURAL NETWORK MODEL

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Main message: Developing optimal flame retardant polymer compositions that meet all aspects of a given application is energy and cost-intensive. To reduce the number of measurements, we developed an artificial neural network-based system to predict the flammability of polymers from small-scale test data and structural properties. Ignition time, peak and total heat release and mass residue after burning reference and flame retarded epoxy resins were predicted.

Keywords: artificial neural network, prediction of flammability, epoxy resin, chemical structure

Introduction

Epoxy resins and their composites are widely used in several industrial sectors (e.g. aerospace and automotive industry) where proper flame retardancy is crucial. Developing novel materials with excellent fire performance is often highly material and cost-intensive and requires several material compositions and destructive tests. To reduce the number of tests, we investigated two methods to predict the flammability of reference and flame retarded (FR) epoxy resins. First, we predicted the total heat release (THR) and the mass residue after burning using calculations based on chemical group contributions according to the work of Lyon et al. [1] and Sonnier et al. [2,3]. We also developed an artificial neural network (ANN)-based system to predict the ignition time, peak and total heat release and mass residue after burning of reference and flame retarded epoxy resins using small-scale thermal and flammability test results and structural properties [4].

Experimental

In both cases, we used our previous experimental results as a database for calculations and prediction. The chemical group contributions were determined according to the literature, and THR and mass residue were calculated in the case of four FR epoxy resin systems: a trifunctional glycerol-based (GER), a tetrafunctional pentaerythritol-based (PER), a sorbitol polyglycidyl ether-based (SPE) and a glucofuranoside triglycidyl ether-based (GFTE) epoxy resin. As hardeners, we used cycloaliphatic amine and diethyl-methylbenzene-diamine (DETDA), while resorcinol bis(diphenyl phosphate) (RDP), N,N',N"-tris(2-aminoethyl)-phosphoric triamide (TEDAP), a phosphorus-containing polyol flame retardant (Clariant OP560) and an alkyl phosphate oligomer (PNX) were used as flame retardants. The samples contained 0, 1, 2, 3, and 4 % phosphorus, respectively. THR and mass residue calculated from the chemical group contributions were compared with the same material compositions' THR and mass residue determined with standard mass loss type cone calorimetry (MLC) tests (100 x 100 x 4 mm samples; 50 kW/m^2 heat flux).

During the development of the model based on artificial neural network (ANN), we focused on small-scale thermal and flammability test results (DSC, TGA, UL-94, LOI) and structural properties. The input parameters included the ratio of carbon (C), hydrogen (H), nitrogen (N), oxygen (O) and phosphorus (P) atoms, the matrix structure (aliphatic, cycloaliphatic or aromatic), the oxygen index, the UL-94 classification, the amount of added flame retardant(s), the type of hardener and matrix, the heat flux used in the MLC measurement. The database included 41 elements (reference and FR epoxy resin compositions) with 15 input and 4 output parameters. We chose the time to ignition (TTI), peak heat release rate (pHRR), total heat release (THR) and mass residue as output parameters. After training the ANN, we predicted the output parameters in GER and PER epoxy resins made with 0, 1, 2, 3, and

4 % phosphorus (P) content from RDP. The prediction results were compared to the MLC results of the same material compositions (validation).

Results and Discussion

The calculations based on group contributions predicted the trends in most cases correctly, e.g. the decrease in THR with increasing P content, up to about 3%. The main reason for the differences between the particular measured and calculated values is that the group contributions were determined by pyrolysis combustion flow calorimetry (PCFC), while we have used MLC results as measured data so far.

The ANN model predicted pHRR values for GER and PER epoxy resins are in good agreement with the test results. (Figure 1). The prediction accuracy of the ANN-based model varied in the following order: THR>pHRR>TTI>mass residue. The relatively larger error in the prediction of TTI and mass residue is due to the inherently larger standard deviation of the test methods. Data outliers in the input database often cause a significant difference between predicted and validated data, but in 65% of cases, the average absolute deviation is below 10%. We used the sensitivity analysis of the output parameters to rank the input parameters according to their impact on the output parameters. The resulting ranking was helpful to establish a relationship between the input and output parameters based on their physical content.

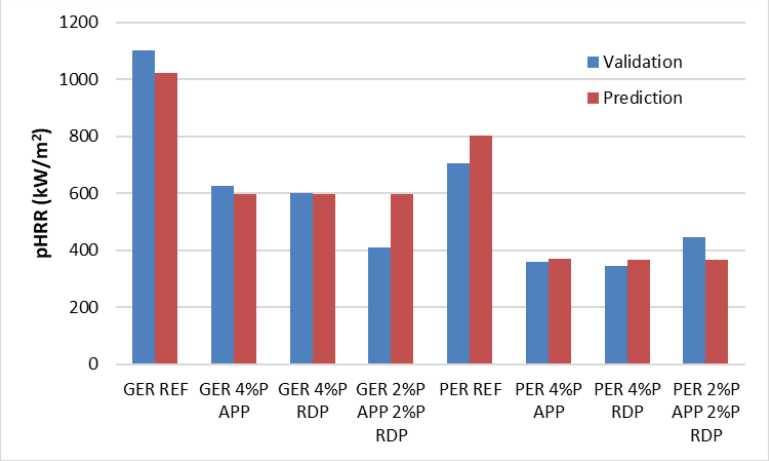

FIGURE 1. Comparison of the measured and predicted pHRR of reference and flame retarded GER and PER epoxy resins

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