DEVELOPMENT OF FIRE RETARDANT EPOXY-BASED GELCOATS FOR CARBON FIBRE REINFORCED EPOXY RESIN COMPOSITES

A. Toldy, Zs. Kovács, Á. Pomázi
Budapest University of Technology and Economics, Department of Polymer Engineering, H-1111 Budapest, Műegyetem rkp. 3.
atoldy@mail.bme.hu

Main message: We prepared carbon fibre reinforced reference and flame retarded epoxy resin composites by wet compression moulding, and investigated the effect of reference and flame retarded gelcoats in two applied thicknesses on their fire performance. If a gelcoat layer is required on the surface of the composite part (e.g. to reach appropriate surface properties), a significant increase in heat release must be taken into consideration, or a flame retarded multifunctional gelcoat should be applied.

Keywords: epoxy resin, carbon fibre reinforced composite, flame retardant gelcoat

Introduction
Achieving special features in polymer composites, such as flame retardancy, often requires the application of different additives, which might negatively affect other properties of the polymer matrix and the composite structure. Furthermore, the application of solid additives in composites produced by liquid transfer moulding can lead to the filtration of the additive by the reinforcement [1], which causes a non-uniform particle distribution and an uneven fire performance. An evident solution to address these issues is to apply the additives in a separate layer on the surface of the composite. As in many applications, gelcoats are used to reach appropriate surface quality, a reasonable progression in the composite industry is the development of multifunctional gelcoats [2].

Experimental
As matrix material we applied low-viscosity, potentially renewable tetrafunctional pentaerythritol-based (PER) epoxy resin (EP) cured with a cycloaliphatic amine-type hardener. Composite laminates were made by wet compression moulding from unidirectional carbon fibre (CF) fabric reinforcement. As reference epoxy gelcoat component we used Sicomin SG715 with Sicomin SD802 hardener, while the titanium-dioxide containing Sicomin SGi128 was used as commercial flame retarded epoxy gelcoat with Sicomin SD228 hardener. For the preparation of flame retarded gelcoats on the basis of Sicomin SG715 we used ammonium polyphosphate (APP) as flame retardant. We prepared gelcoat formulations with 5%, 10% and 15% P-content. For the flame retardancy of the composite matrix material we used resorcinol bis(diphenyl phosphate) (RDP).
The thermal stability of the matrices was investigated by thermogravimetric analysis, while the effect of FRs on the glass transition temperature and crosslinking process was studied by differential scanning calorimetry. Fire retardancy of gelcoat and coated composite specimens was evaluated by limiting oxygen index (LOI), UL-94 tests and mass loss calorimetry.

Results and Discussion
We compared the glass transition temperature (T_g), reaction enthalpy of crosslinking, thermal stability and fire performance of epoxy resin-based gelcoats prepared with 5, 10 and 15% P-content from APP to the reference epoxy resin-based gelcoat and a commercially available, flame retarded epoxy resin-based gelcoat containing titanium dioxide. The well-dispersed APP particles increased the T_g of the gelcoats, and the reaction enthalpy of crosslinking related to epoxy parts increased. All APP-containing gelcoats overperformed the commercially available reference and FR gelcoat in terms of thermal stability. By increasing the APP concentration, both the thermal stability and the limiting oxygen index (LOI) of the gelcoats increased. All FR gelcoats reached the self-extinguishing, V-0 UL-94 classification.

The 4 mm thick gelcoat specimens foamed into the cone during the mass loss calorimetry (MLC) even at 25 kW/m^2; therefore, we repeated the tests with 2 mm thick gelcoats. As for the heat release rates, 5%P APP content was not enough to outreach the commercial SGi128 FR gelcoat. The gelcoat with 10%P APP behaved similarly, while the formulation with 15%P APP outperformed the SGi128 FR gelcoat.
We coated the reference and flame retarded composite substrates in 0.5 and 1 mm thicknesses with the commercial reference and flame retarded gelcoats, as well as with the APP-containing ones. According to the mass loss calorimetry results, gelcoats containing no flame retardants significantly impaired the fire performance of the fibre reinforced composites, leading to high peak and total heat release rates, even when applied in 0.5 mm thickness. In average, the 0.5 mm thick FR gelcoats lowered the pHRR of the PER composite by 22%, which could be further decreased by 19% with the addition of RDP to the composite matrix. Among the composites coated with 0.5 mm thick gelcoat, the lowest pHRR (180 kW/m^2) was reached by the FR composite coated with 15% P-containing gelcoat (SG715 15%P APP).
The 1 mm thick FR gelcoats (Fig. 1) lowered the pHRR of the PER composite in average twice as much the 0.5 mm thick coatings, however the additional FR effect of the RDP-incorporation was lowered at the same time. The same pHRR (180 kW/m^2) was reached by the PER composite coated with 1 mm thick 15% P-containing gelcoat (SG715 15%P APP) without FR in the matrix, as with the composite containing RDP in the matrix in combination with a 0.5 mm thick 15% P-containing gelcoat. Among all composites, the lowest pHRR (149 kW/m^2) was reached by the FR composite coated with 15% P-containing gelcoat (SG715 15%P APP), which is equivalent to a pHRR reduction of 58% compared to the uncoated PER reference composite, and 67% reduction compared to the PER reference composite coated with 1 mm thick non-FR gelcoat, respectively.

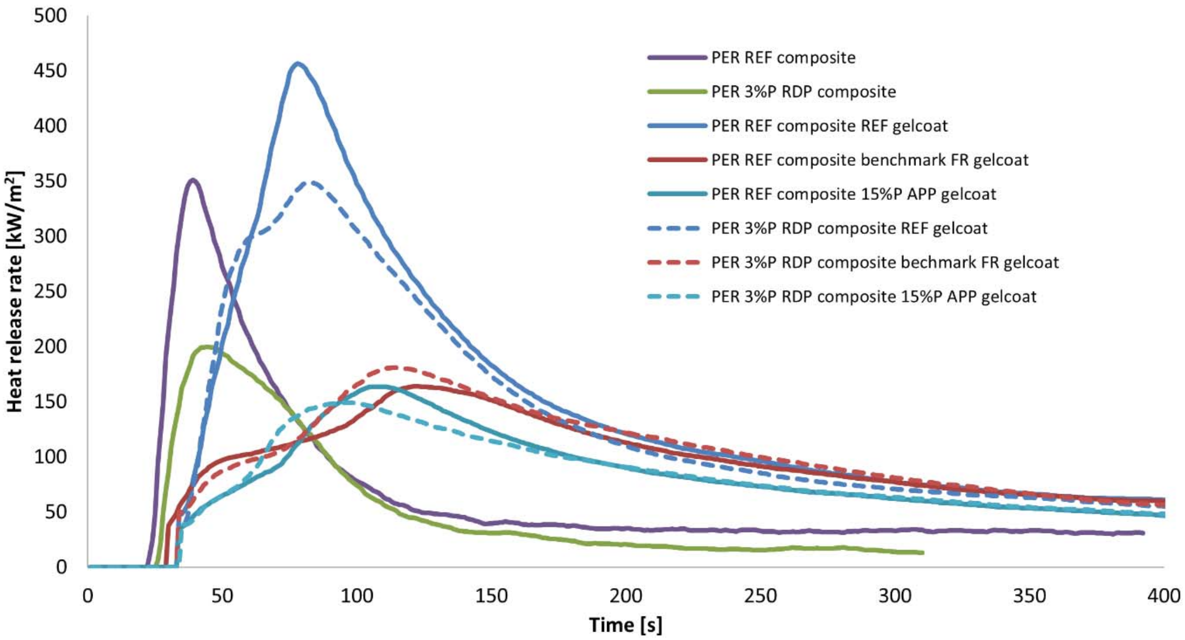

FIGURE 1. Heat release rate of reference and FR composites coated with 1 mm thick gelcoats

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