Mechanical properties of reactively flame retarded cyanate ester/epoxy resin blends and their carbon fibre reinforced composites

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Abstract. Cyanate ester/epoxy resin (CE/EP) carbon fibre reinforced composites consisting of diglycidyl ether of bisphenol A (DGEBA) and novolac type cyanate ester (CE) were prepared and reactively flame retarded using epoxy functional adduct of DGEBA and 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO). Effect of cyanate ester and flame retardant (FR) ratio was determined on matrix viscosity, matrix and composite glass transition temperature (T_g), as well as composite mechanical properties including storage modulus, tensile, bending, interlaminar shear and Charpy impact properties. Although the epoxy resin (EP) and FR decreased the T_g, even the flame retarded CE/EP blends had at least 22 °C higher T_g than the benchmark DGEBA composite. As for the mechanical properties, as a result of higher interlaminar shear strength suggesting better fibre-matrix adhesion, the CE/EP blends managed to over-perform the reference CE in most cases: The 2% phosphorus (P)-containing CE/EP composite had 25% higher tensile strength than the CE reference. The bending strength of the blends remained in the same range as the reference, while the impact resistance significantly increased in comparison to CE, especially in flame retarded composites.

Keywords: mechanical properties, flame retardancy, cyanate ester, epoxy resin, carbon fibre reinforced composites

1. Introduction

Even in case of high performance thermosetting polymer materials as cyanate esters and epoxy resins the flame retardancy is still an issue to be solved, particularly in advanced sectors with strict safety requirements as electrical and aeronautical industry. However, the addition of flame retardants usually decreases the glass transition temperature and mechanical properties of the polymers [1]. One possibility to minimize these effects is to apply reactive flame retardants, which can be chemically incorporated into the polymer structure. This approach offers further advantages: as the flame retardant does not migrate to the surface of the matrix either during high temperature processing or application, it provides more stable effect compared to additive flame retardants and lower ratio is sufficient to achieve the same level of flame retardancy [2]. The increasing focus on the health and environmental compatibility of flame retardants also facilitated the headway of this reactive approach, in particular many organophosphorus reactive flame retardants were developed in the recent years [3–6]. Another way to compensate the effect of flame retardants is to form blends with another polymer possessing high glass transition temperature, thermally stable backbone and outstanding me-

chanical properties. Again, instead of simple blend formation reactive blending resulting in primary chemical bonds between the polymers is preferred. For these reasons epoxy resins are often blended with cyanate esters [7–10], which can be applied instead of the commonly applied amine or anhydride type hardeners. This way cyanate esters can be used as multifunctional reactive modifiers increasing the glass transition temperature, and improving the thermal stability and mechanical properties of flame retarded epoxy resins.

In CE/EP systems either the EP [11, 12] or the CE component [13–15] can contain the phosphorus (P) -containing flame retardant unit, but separate P-containing reactive modifiers (mostly with –OH [16] and –NH$_2$ functions [17]) can be applied as well. These articles studied the flame retardancy results of P-containing CE/EP or CE, along with the effect on glass transition temperature and in some cases on modulus only in polymer matrices. To the extent of our knowledge, no articles have been published yet on the effects of phosphorus flame retardants on glass transition temperature and mechanical properties in CE/EP fibre reinforced composites, therefore our current study aims at filling in this gap.

In particular, in this work the EP component, DGEBA was pre-reacted with DOPO in order to obtain an epoxy functional reactive flame retardant, and a novolac type, high glass transition temperature CE was blended with it. As the flame retardants generally decrease the glass transition temperature of EPs, the hybrid system consisting of CE, EP and reactive flame retardant was made in order to reach higher glass transition temperature than in case of flame retarded EP alone.

Based on flame retardancy results of these CE/EP systems (published previously by the authors [18]), the best performing blends were chosen, and the effect of CE and flame retardant ratio was determined on viscosity, glass transition temperature (T_g) and dynamic mechanical properties. From these best performing blends also reactively flame retarded CE/EP carbon fibre reinforced composites were made and their dynamic mechanical, tensile, bending, interlaminar shear strength and Charpy impact properties were tested and compared to CE and EP benchmarks.

2. Materials and methods
2.1. Materials
Novolac type cyanate ester (Primaset PT-30) was acquired from Lonza Ltd. (Basel, Switzerland).

Diglycidyl ether of bisphenol A (DGEBA, Ipox ER 1010) with 188 g/eq epoxy equivalent weight was obtained from IPOX Chemicals Ltd. (Budapest, Hungary).

As reactive flame retardant 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO, Struktol Polydis 3710, properties: molecular mass: 216.17 g/mol, melting point: 116 °C) was used supplied by Struktol Gmbh (Hamburg, Germany).

In order to form a phosphorus-containing epoxy component, DOPO was reacted with DGEBA in 1:1 molar ratio. Before the reaction DOPO was kept at 85 °C for 12 h, in order to eliminate the traces of moisture. DGEBA was kept under vacuum at 110 °C to remove air and traces of moisture, and after adding DOPO, the mixture was stirred at 160 °C for 5 h. After cooling to room temperature a solid adduct was obtained. This synthesis, based on the method of Wang and Lin [19], was published by the authors previously [18]. The main advantage of the adduct formation is, that this way the highly intensive reaction between DOPO and PT-30, furthermore carbamate and consequent CO$_2$ formation from CE due to water traces present in DOPO (despite careful drying) can be avoided. Due to controlled reaction conditions and stoichiometry, an oxirane functional adduct is formed, which reacts the same way as DGEBA with PT-30 (main reactions: trimerization of cyanates, insertion of oxiranes into the cyanurate, isomerization of alkyl-substituted triazines and formation of oxalidinones from isocyanurates and glycidyl ethers [20]).

As hardener methyl-tetrahydrophthalic-anhydride (Aradur 917 – AR917) was applied with 1-methylimidazole (DY070) accelerator by Huntsman Advanced Materials (Basel, Switzerland). The equivalent mass of the anhydride type curing agent, calculated from its molecular mass, was 160 g/eq. The accelerator was applied in 2 mass% related to the mass of DGEBA. The chemical structures of the used polymer components are shown in Figure 1.

As fibre reinforcement Zoltek Panex 35 type unidirectional carbon weave with 300 g/m^2 areal weight provided by Zoltek Ltd. (Nyergesújfalu, Hungary) was applied.
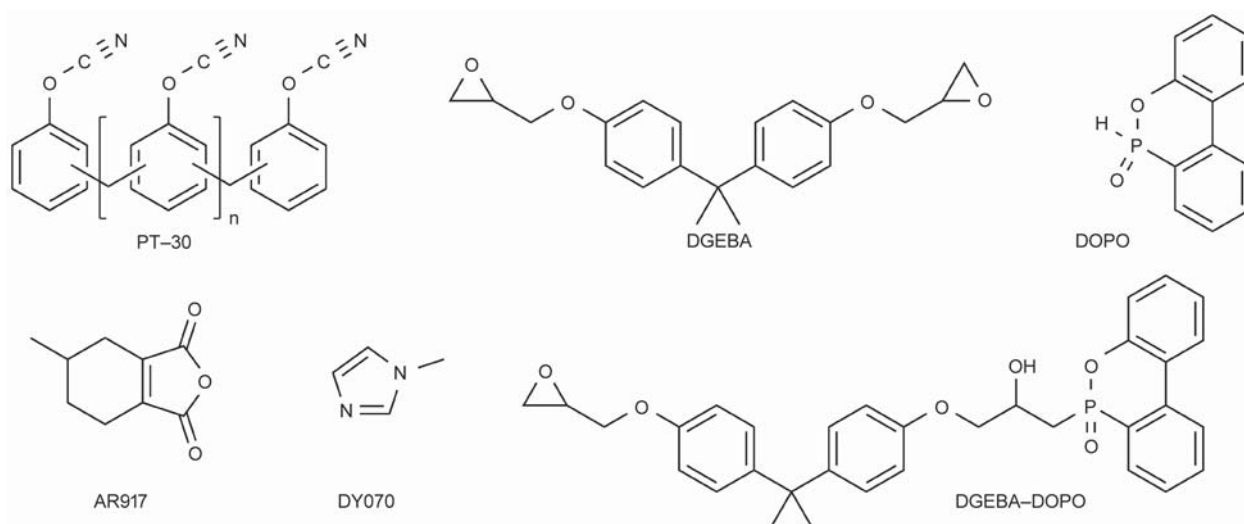
Figure 1. Chemical structures of the used polymer components

2.2. Methods

2.2.1. Sample preparation

Besides the reference CE, EP and CE/EP matrices, flame retarded CE/EP matrices with 2 and 3 mass% phosphorus were prepared using the synthesized DGEBA – DOPO adduct. The composition of the composite matrices is summarized in Table 1.

Polymer matrix specimens for the mechanical investigations were made by resin moulding with a vertical moulding tool. For the preparation of matrix specimens for flame retardancy testing heat resistant silicone moulds were used of appropriate size. The composite laminates were made by hand lamination followed by press moulding. Each carbon weave layer was separately impregnated, in case of high viscosity matrices the polymer and the mould were heated to 80 °C. The prepared laminates were put under compression with 25 bar pressure in T30 type platen press (Metal Fluid Engineering s. r. l., Verdello Zingonia, Italy) to achieve high and uniform fibre content in the composites. For flame retardancy measurements 4 mm thick specimens were made using $[0]_{10}$ of carbon weave, while for mechanical tests 2 mm thick laminates were made with $[0]_5$ layup. The heat treatment was carried out during pressing. Samples containing PT-30 were cured 1 h at 150 °C, 3 h at 200 °C and 1 h 260 °C according to the suggestion of CE supplier. In case of DGEBA the heat treatment consisted of a 1 h 80 °C and 3 h 140 °C isothermal step. The measured fibre content of the composites was in the range of 50–55 mass%.

2.2.2. Parallel plate rheometry

Viscosity was determined by parallel plate rheometry using AR2000 device from TA Instruments (New Castle, DE, USA) in the range of 25–80 °C, with 5 °C/min temperature ramp, applying 40 mm diameter plate and 150 μm gap between the plates.

2.2.3. Flame retardant characterization

The fire behaviour of the reference and flame retarded systems was characterized by limiting oxygen

Table 1. Composition of the CE, EP and CE/EP composite matrices

	Sample composition [mass%]				
	PT-30	**DGEBA**	**DOPO**	**AR917**	**DY070**
Reference samples					
PT-30	100	–	–	–	–
DGEBA	–	52.3	–	47.1	0.6
20% PT-30 - 80% DGEBA	20	80	–	–	–
40% PT-30 - 60% DGEBA	40	60	–	–	–
Flame retarded samples					
40% PT-30 - DGEBA – DOPO 2% P	40	46.1	13.9	–	–
40% PT-30 - DGEBA – DOPO 3% P	40	39.1	20.9	–	–

index measurements (LOI, according to ASTM D-2863). The LOI value expresses the minimum volume fraction of oxygen in a mixture of oxygen and nitrogen that supports flaming combustion of a material under specified test conditions.

Standard UL-94 flammability tests (according to ASTM D3081 and ASTM D-635, respectively) were also carried out in order to classify the samples based on their flammability in horizontal and vertical test setups.

Mass loss calorimeter tests were carried out by an instrument made by FTT Inc. (East Grinstead, UK) according to ISO 13927 standard method. Specimens (100×100×4 mm) were exposed to a constant heat flux of 50 kW/m^2 and ignited. Heat release values and mass reduction were continuously recorded during burning.

2.2.4. Dynamic mechanical analysis (DMA)

For the investigations of the dynamic mechanical properties and for the determination of the glass transition temperature (T_g) values DMA tests were carried out in three point bending setup with TA Q800 device of TA Instruments (New Castle, DE, USA). The temperature range was 0–260 °C (in case of pure CE samples 0–400 °C) with 3 °C/min heat rate. The frequency was 1 Hz. The size of the specimens was 55×10×2 mm (length × width × thickness), and the support span was 50 mm. The amplitude was strain controlled with 0.1% relative strain. From the results glass transition temperature based on the tanδ peaks (T_g) and storage modulus (E') values at 25 and 75 °C were determined by the software of the DMA device (TA Instruments Universal Analysis 2000 4.7A version).

2.2.5. Tensile test

Tensile tests were carried out to determine the composites tensile strength and tensile modulus values (E_m) by a Zwick Z250 (Ulm, Germany) type computer controlled universal tester, equipped with a 20 kN capacity load cell. Based on EN ISO 527 the specimen size was 140×10×2 mm (length × width × thickness). The test speed was 2 mm/min, and the initial test length was 80 mm. During the test, force and displacement values were recorded and the tensile parameters were calculated according to the standard. In each case 5 parallel tests were carried out.

2.2.6. Bending test

Bending tests were carried out in three point bending setup to determine the composites flexural strength and flexural modulus values by a Zwick Z250 (Ulm, Germany) type computer controlled universal tester, equipped with a 20 kN capacity load cell with standard three point bending fixtures. The size of the specimens, based on EN ISO 14125 was 100×10×2 mm (length × width × thickness). The test speed was 5 mm/min, and the span length was 80 mm. During the test, force and deflection values were recorded and the bending parameters were calculated according to the standard. In each case 5 parallel tests were carried out.

2.2.7. Interlaminar shear test

According to EN ISO 14130 interlaminar shear tests were carried out on 5–5 specimens with 20×10× 2 mm size (length x width x thickness) by a Zwick Z020 (Ulm, Germany) universal tester. The support span was 10 mm and the test speed was 1 mm/min. From the registered force-deflection results apparent interlaminar shear strength was calculated and compared.

2.2.8. Charpy impact test

Charpy impact tests were carried out according to EN ISO 179-1 by a normal impact on unnotched specimens of 80 mm length, 10 mm width and 2 mm thick with a Ceast Resil Impactor Junior (Torino, Italy) instrumented pendulum equipped with a 2 J hammer using 2.9 m/s impact velocity, with 150° starting angle and 62 mm support span on 5–5 specimens from each sample. The force–time curves were registered by a Ceast DAS 8000 data acquisition unit and the Charpy impact energy was calculated and compared.

3. Results and discussion
3.1. Viscosity of polymer matrices

One major aspect of the processing of resin systems is their viscosity, therefore prior to composite preparation viscosity of the polymer matrices was determined in the function of temperature. According to Hay [21] for resin injection 100–300 mPa·s, for pultrusion 400–800 mPa·s, while for filament winding viscosity of 800–2000 mPa·s is recommended. Cyanate esters are often processed by filament winding, where the filaments are immersed into a heat-
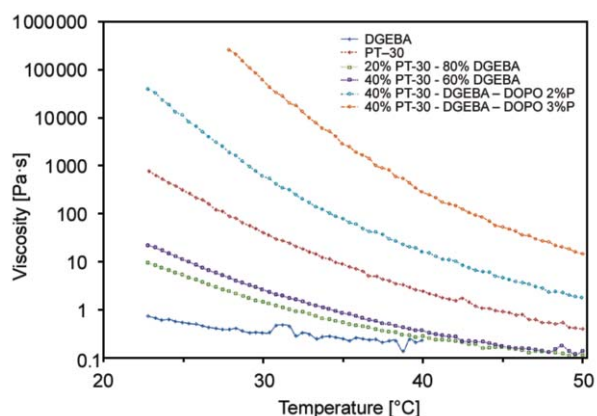
Figure 2. Viscosity of the CE and EP references and CE/EP blends in the temperature range of 25–80 °C

Table 2. Viscosity of the CE and EP references and CE/EP blends at 80 °C

Sample	Viscosity [mPa·s]
PT-30	400
DGEBA	233[*]
20% PT-30 - 80% DGEBA	107
40% PT-30 - 60% DGEBA	113
40% PT-30 - DGEBA – DOPO 2%P	1 623
40% PT-30 - DGEBA – DOPO 3%P	14 780

[*]at 60 °C-on due to lower gel time

able resin bath allowing the reduction of the matrix viscosity by increasing its temperature. The viscosity of the CE and EP references and CE/EP blends in the temperature range of 25–80 °C can be seen in Figure 2.

By increasing the amount of CE in the blends, the viscosity increased, as expected. Furthermore, the addition of solid DOPO-DGEBA adduct significantly increased the viscosity as well. By increasing the temperature the viscosity of the matrices showed a monotone decreasing tendency.

According to the viscosity values at 80 °C (Table 2) the samples containing 3 mass% phosphorus can be rather processed by hot pressing, Blends containing 2 mass% phosphorus are suitable for filament winding as well.

Based on these results hand lamination followed by hot pressing was chosen as composite preparation method, as it provides high fibre content and excellent reproducibility.

3.2. Flame retardancy of polymer matrices and composites

In order to be able to judge the overall performance of the CE/EP systems, their LOI, UL-94 and most important mass loss calorimetry results were summarized in Table 3. The flame retardancy results of the CE/EP matrices itself along with the results of polymer composites made thereof are discussed in detail elsewhere [18].

The addition of CE to EP significantly increased the LOI value, however it was not sufficient to improve the HB UL-94 rate of the samples. All blends consisting of EP, CE and phosphorus-containing flame retardant reached the V-0 UL-94 classification and

Table 3. LOI, UL-94 and mass loss calorimetry results of the composites made of CE and EP references and their blends

	LOI [V/V%]	UL-94[*]	TTI [s]	pHRR [kW/m²]	THR [MJ/m²]	Residue [mass%]
Matrix sample						
PT-30	30	HB	26	156	15.5	48
DGEBA	23	HB (17.1±2)	40	743	91.0	0
20% PT-30 - 80% DGEBA	33	HB	50	471	59.6	0
40% PT-30 - 60% DGEBA	28	HB	50	238	55.1	14
40% PT-30 - DGEBA – DOPO 2%P	43	V-0	53	195	36.3	23
40% PT-30 - DGEBA – DOPO 3%P	45	V-0	44	234	47.5	22
Composite sample						
PT-30	58	V-0	80	84	9.8	81
DGEBA	33	HB	55	176	37.9	50
20% PT-30 - 80% DGEBA	41	HB	51	162	29.9	61
40% PT-30 - 60% DGEBA	42	V-0	87	134	21.8	70
40% PT-30 - DGEBA – DOPO 2%P	46	V-0	72	101	20.1	67
40% PT-30 - DGEBA – DOPO 3%P	48	V-0	70	84	18.7	67

[*]in parenthesis the horizontal burning rate is showed, where measurable
LOI: limiting oxygen index, TTI: time to ignition, pHRR: peak of heat release rate, THR: total heat released
Flame retarded samples are highlighted in grey (2%P) and dark grey (3%P)

exhibited intensive intumescent charring and increased residual mass. Although the carbon fibre reinforcement plies hinder the solid phase intumescent mechanism of the phosphorus flame retardant [22], as the reinforcement itself is inflammable under the conditions of the flame retardancy tests, the composite specimens showed even better flame retardant properties than the polymer matrices alone. In composite specimens 40 mass% of PT-30 alone was sufficient to reach the V-0 UL-94 rate. By increasing the amount of CE and DOPO, the LOI increased and the pHRR values showed further decrease. The 40% PT-30 - DGEBA – DOPO 3%P composite had the same pHRR value, 84 kW/m^2 as the PT-30 reference composite.

3.3. Dynamic mechanical analysis of polymer matrices and composites

Storage modulus curves of the CE and EP references and CE/EP blends are displayed in Figure 3, while that of the composites can be seen in Figure 4. Glass transition temperature based on the tanδ peaks (T_g)
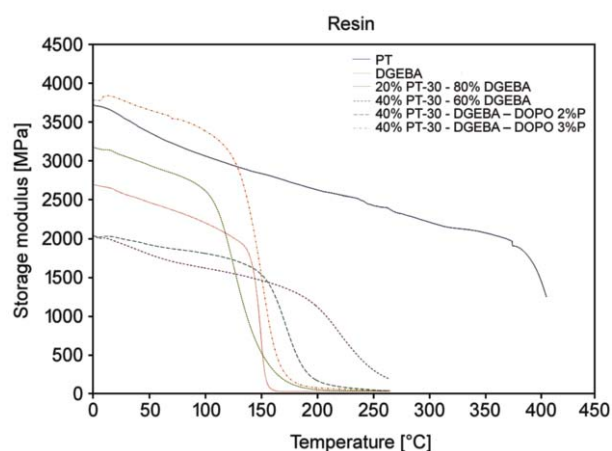
Figure 3. Storage modulus of the CE and EP references and CE/EP blends in the temperature range of 25–260 °C (in case of pure CE 25–400 °C)
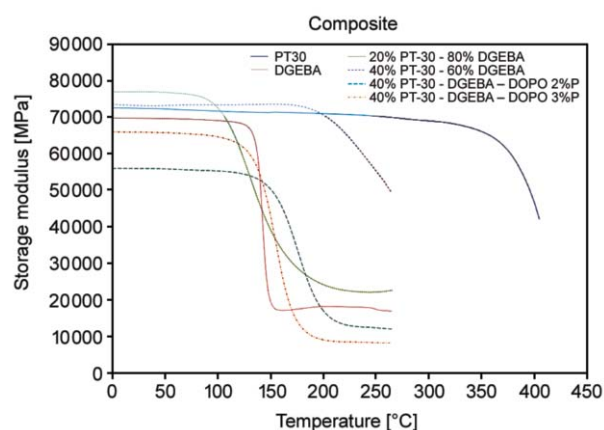
Figure 4. Storage modulus of the CE and EP reference and CE/EP blend composites in the temperature range of 25–260 °C (in case of pure CE 25–400 °C)

and storage modulus (E') values at 25 and 75 °C are shown in Table 4.

In case of resin samples by increasing the temperature the storage moduli showed a decreasing tendency, while in case of composite samples it remained in the same range at least up to 75 °C. As for the CE/EP resin blends, the 20% PT-30 - 80% DGEBA had higher storage modulus up to 115 °C, while the 40% PT-30 - 60% DGEBA blend showed better properties than DGEBA only above 140 °C, similarly to its flame retarded version with 2%P. However, the 40% PT-30 - DGEBA – DOPO 3%P matrix sample performed better than DGEBA in the whole temperature range, and had even higher storage modulus than CE up to 125 °C, which may be explained by the relative stoichiometric excess of PT-30 (related to the amount of oxirane groups present in DGEBA and DOPO-DGEBA components in the sample). In case of composite samples the 20% PT-30 - 80% DGEBA blend had higher storage modulus than CE up to 90 °C and higher than DGEBA up to 105 °C. The 40% PT-30 - 60% DGEBA composite per-

Table 4. Glass transition temperature (T_g) and storage modulus values at 25 and 75 °C of CE/EP matrices and composites determined by DMA

Sample	Glass transition temperature [°C]		Storage modulus at 25 °C [MPa]		Storage modulus at 75 °C [MPa]	
	Matrix	Composite	Matrix	Composite	Matrix	Composite
PT-30	401	394	3572	72 407	3196	71 908
DGEBA	155	145	2585	69 691	2343	69 407
20% PT-30 - 80% DGEBA	172	145	3071	92 311	2815	91 420
40% PT-30 - 60% DGEBA	247	249	1932	73 150	1686	73 360
40% PT-30 - DGEBA – DOPO 2%P	188	187	1995	55 967	1856	55 537
40% PT-30 - DGEBA – DOPO 3%P	165	167	3784	65 882	3534	65 378

formed similarly as CE up to 200 °C and outperformed DGEBA in the whole temperature range. The flame retarded composites showed somewhat lower storage modulus than DGEBA (except the 140–165 °C range in case of 40% PT-30 - DGEBA – DOPO 3%P composite, and the 140–190 °C range in case of 40% PT-30 - DGEBA – DOPO 2%P), most probably due to lower fibre-matrix adhesion (see the interlaminar shear properties in chapter 3.4.).

As for the glass transition temperatures (Table 4), in case of resin samples the T_g of the blends increased with increasing amount of CE. Compared to 40% PT-30 - 60% DGEBA sample, the inclusion of flame retardants decreased the T_g, most probably due to lower crosslinking density, however it was still above the T_g of DGEBA. In case of composite samples the T_g decreased in CE and EP reference samples and in 20% PT-30 - 80% DGEBA sample, while in 40% PT-30 containing composites, including the flame retarded ones, practically it remained the same value as in case of the matrix samples. Compared to DGEBA, the 40% PT-30 - DGEBA – DOPO 2%P composite showed 42 °C increase, while the 40% PT-30 - DGEBA – DOPO 3%P composite had still 22 °C higher T_g.

3.4. Tensile, bending, interlaminar shear and Charpy impact properties of polymer composites

Tensile properties of the CE and EP reference composites and CE/EP blend composites are shown in Table 5. According to the tensile test results, the inclusion of EP significantly increased the tensile strength of the rigid CE. More surprisingly, by adding DOPO-DGEBA adduct to the 40% PT-30 - 60% DGEBA matrix, the tensile strength of the 2% P-containing composite increased even further, and in case of 3% P-containing sample it still remained over the value of the CE reference. This amelioration may be attributed to better fibre-matrix adhesion and to the reactive nature of the flame retardant: by incorporating it by primary chemical bonds to the matrix itself, it does not migrate to the matrix surface either during high temperature processing or application. The strain at break increased to some extent in all blends containing DGEBA in comparison to the reference CE, decreasing the rigidity of it. The highest tensile modulus was reached in case of 20% PT-30 - 80% DGEBA, higher than the moduli of the blend components themselves. By adding DOPO-DGEBA adduct to the system, the tensile modulus slightly decreased.

Flexural properties of the CE and EP reference composites and CE/EP blend composites are shown in Table 6. According to the results the addition of EP into CE resulted in slightly higher flexural strength than in case of the reference CE and EP itself. The inclusion of DOPO-DGEBA adduct decreased the flexural strength and modulus, and increased the deformation at break, however taking into account the standard deviation values, the flexural strength and modulus of 40% PT-30 - DGEBA – DOPO 3% re-

Table 5. Tensile properties of the CE and EP reference composites and CE/EP blend composites

Composite sample	Tensile strength [MPa]	Strain at break [%]	Tensile modulus [GPa]
PT-30	689.2±100.9	4.43±0.60	27.7±0.7
DGEBA	912.6±45.7	5.35±0.43	26.8±2.4
20% PT-30 - 80% DGEBA	1040.9±43.0	5.66±0.22	28.8±0.2
40% PT-30 - 60% DGEBA	844.1±40.3	5.06±0.16	25.1±2.1
40% PT-30 - DGEBA – DOPO 2%P	861.2±54.7	5.73±0.47	24.9±0.4
40% PT-30 - DGEBA – DOPO 3%P	715.2±32.4	5.06±0.19	23.4±0.2

Table 6. Flexural properties of the CE and EP reference composites and CE/EP blend composites

Composite sample	Flexural strength [MPa]	Deformation at break [%]	Flexural modulus [GPa]
PT-30	1227.0±271.1	1.36±0.03	103.2±19.5
DGEBA	1203.0±115.9	1.36±0.09	98.2±4.3
20% PT-30 - 80% DGEBA	1240.1±114.3	1.36±0.04	100.1±10.9
40% PT-30 - 60% DGEBA	1238.5±79.2	1.37±0.04	98.2±8.8
40% PT-30 - DGEBA – DOPO 2%P	1056.2±54.1	1.43±0.02	79.5±5.0
40% PT-30 - DGEBA – DOPO 3%P	1149.0±96.7	1.45±0.09	96.0±10.9

mained in the same range as in case of CE and EP references.

In order to find an explanation for the tensile and flexural properties, the interlaminar shear strength of the composites was determined (Table 7). In accordance with the tensile and bending properties, the interlaminar shear strength values of the CE/EP blends were higher than in case of the CE and EP references. The inclusion of the polar phosphorus-containing flame retardant decreased the interlaminar shear strength, however these values were still well above the value of the reference CE composite.

Charpy impact test is suitable for comparing the impact resistance of the composites. The results of the instrumented Charpy unnotched impact measurements are given in Table 8. The impact strength of the 20% PT-30 - 80% DGEBA blend was practically the same as in case of DGEBA, however, the 40% PT-30 - 60% DGEBA blend had even higher impact strength than CE. By increasing the amount of flame retardants, the fracture toughness showed further in-

crease in comparison to CE, meaning that the flame retarded composites are less brittle than the CE/EP blends and CE, EP references.

4. Conclusions

Reactively flame retarded cyanate ester/epoxy resin (CE/EP) carbon fibre reinforced composites consisting of diglycidyl ether of bisphenol A (DGEBA), novolac type cyanate ester (CE) and an epoxy functional adduct of DGEBA and 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO) were prepared and tested. Influence of cyanate ester and flame retardant addition was determined on matrix viscosity, matrix and composite glass transition temperature (T_g), as well as composite mechanical properties.

From the tested CE/EP composites, the overall performance of V-0 UL-94 rated ones is summarized in Table 9. As expected, the T_g of the CE composite was the highest, however even the flame retarded CE/EP blends had at least 22 °C higher Tg than the benchmark DGEBA composite. As for the mechanical properties, the CE/EP blends outperformed the CE composite in most cases: The addition of EP considerably increased the tensile strength of the rigid CE, more unexpectedly, by adding DOPO-DGEBA adduct to the 40% PT-30 - 60% DGEBA matrix, the tensile strength of the 2% P-containing composite reached a 25% increase compared to the CE composite. The inclusion of EP into CE also resulted in slightly higher flexural strength than in case of the reference CE and EP itself, which was somewhat decreased by the DOPO-DGEBA adduct. These results may be interpreted by the better fibre-matrix adhesion: the interlaminar shear strength of the 40% PT-30 - 60% DGEBA blend was 70% higher than in case of the CE reference. Although the inclusion of the polar phosphorus-containing flame retardant decreased the interlaminar shear strength, their values were still high above the value of the CE composite. The impact strength also increased in comparison to

Table 7. Interlaminar shear strength of the CE and EP reference composites and CE/EP blend composites

Sample	Interlaminar shear strength [MPa]
PT-30	40.0±1.3
DGEBA	61.3±1.9
20% PT-30 - 80% DGEBA	66.8±3.1
40% PT-30 - 60% DGEBA	68.3±3.6
40% PT-30 - DGEBA – DOPO 2%P	53.4±2.0
40% PT-30 - DGEBA – DOPO 3%P	47.9±2.1

Table 8. Charpy impact strength of the CE and EP reference composites and CE/EP blend composites

Sample	Charpy impact strength [J/mm²]
PT-30	90.1±8.0
DGEBA	84.3±5.2
20% PT-30 - 80% DGEBA	84.6±2.9
40% PT-30 - 60% DGEBA	98.3±32.0
40% PT-30 - DGEBA – DOPO 2%P	99.1±15.1
40% PT-30 - DGEBA – DOPO 3%P	113.7±14.0

Table 9. Overall performance of UL-94 V-0 rated CE/EP composite samples (best achieved values are highlighted in bold)

Composite sample	Glass transition temperature [°C]	Tensile strength [MPa]	Flexural strength [MPa]	Interlaminar shear strength [MPa]	Charpy impact strength [J/mm²]
PT-30	**394**	689.2±100.9	1227.0±271.1	40.0±1.3	90.1±8.0
40% PT-30 - 60% DGEBA	249	844.1±40.3	**1238.5±79.2**	**68.3±3.6**	98.3±32.0
40% PT-30 - DGEBA – DOPO 2%P	187	**861.2±54.7**	1056.2±54.1	53.4±2.0	99.1±15.1
40% PT-30 - DGEBA – DOPO 3%P	167	715.2±32.4	1149.0±96.7	47.9±2.1	**113.7±14.0**

CE, from all composites the flame retarded ones were the less brittle.

Acknowledgements

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