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Effect of novel intumescent flame retardant on mechanical and flame retardant properties of continuous glass fibre reinforced polypropylene composites

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ABSTRACT

In this study, the influence of a novel intumescent flame retardant (IFR) on the mode I interlaminar fracture toughness and flame retardant properties of continuous glass fibre reinforced polypropylene (CGF/PP) composites is presented. The novel IFR system that consists of ammonium polyphosphate/poly (ethanediamine-1, 3, 5 triazine-o-4-amion-2, 2, 6, 6- tetramentylniperidine) (APP/PETAT) was introduced into the CGF/PP unidirectional prepregs (UD) composites by hot-melt impregnation process. Double cantilever beam (DCB) tests were performed to evaluate the mode I interlaminar fracture toughness (G_{IC}) by using modified beam theory (MBT). It was found from the experimental results that compared to the neat CGF/PP composite, the mode I fracture toughness initiation toughness ($G_{IC\ init.}$) and propagation toughness ($G_{IC\ prop.}$) of CGF/PP/12.5 wt% IFR composites were increased by an average of 99.7% and 119.11% respectively. Meanwhile, samples with 12.5 wt % IFR could self-extinguish with an oxygen concentration of 32.4% and exhibit lower fire spread speed in UL-94 test. Furthermore, the peak of heat release rate (PHRR), total heat release (THR), fire performance index (FPI), and mass loss rate (MLR) tests indicated that IFR could dramatically enhance the flame retardancy of the composites.

1. Introduction

Despite comparatively difficult preimpregnated due to the high viscosity of resins, fibre-reinforced thermoplastic composites have quickly gained interest and are highly attractive for military and civil infrastructure applications [1–3]. Of the continuous glass fibre (CGF) reinforced thermoplastic composites, polypropylene (PP) is a widely used thermoplastic resin because of its low density, low cost, and high chemical resistance. However, continuous glass fibre reinforced polypropylene (CGF/PP) composites possess some disadvantages, such as poor fire resistance, which limits their use in wider applications [2,4,5]. Although the influence of the mechanical and the flame-retardant properties of PP resin has been widely investigated, there are few studies on the current properties of CGF/PP composites [6–8]. Current research shows that the flame retardant mechanism of resin is not suitable for fibre reinforced polymer matrix composites due to the wicking actions caused by continuous glass fibre [5,9].

Since traditional halogen flame retardants produce toxic gas and smoke, which are environmentally unfriendly, intumescent flame retardants (IFR) provide an efficient way to solve this problem [10–13]. As one of the most commonly and widely reported IFRs, the ammonium polyphosphate / melamine / pentaerythritol (APP/MEL/PER) system can enhance the flame retardancy property of PP resin. The flame retardancy mechanism of IFR is the formation of continuous and swollen char on the surface of the resin when the surface temperature reaches a critical value, which acts as an insulative barrier between the fire and resin substrate. However, the APP/MEL/PER IFR systems are mainly composed of small molecule compounds, which are moisture sensitive, leading to a deterioration of the flame retardancy [6]. In our recent work [14], a novel macromolecular charring agent poly(ethanediamine-1,3,5-triazine-p-4-amino-2,2,6,6-tetramethylpiperidine) (PETAT) was designed and synthesized, and it showed an excellent synergism with APP in flame retardant PP resin. The corresponding chemical structure is shown in Fig. 1.

The improvements in terms of flame retardancy property with flame retardant adding are usually accompanied with the changing of the

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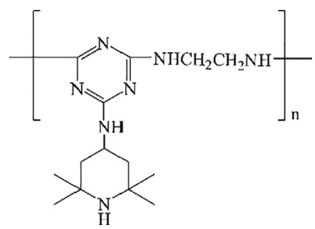

Fig. 1. Chemical structure of PETAT.

Table 1
Designation and composition of CGF/PP/IFR UD.

Samples	PP (wt %)	CGF (wt %)	APP (wt %)	PETAT (wt %)	MAPP (wt %)
CGF/PP/0 wt% IFR	47	50	0	0	3
CGF/PP/4.5 wt% IFR	42.5	50	3	1.5	3
CGF/PP/8.5 wt% IFR	38.5	50	5.7	2.8	3
CGF/PP/12.5 wt% IFR	34.5	50	8.3	4.2	3
CGF/PP/16.5 wt% IFR	30.5	50	11	5.5	3

mechanical properties of composites. Interlaminar failure or delamination is one of the most critical failure mode in composite laminates. Extensive investigations have been carried out to research various factors on the interlaminar fracture toughness of composites, such as ply thickness [15], interface fibre angle [16], crack velocity [17], and nanomatearials [7,18,19]. Among those researches, very limited studies about the influence of interfacial adhesion between fibre and matrix on the interlaminar fracture toughness of composites could be found. In general, engineering the interface adhesion between fibre and matrix is an important technique to enhance the interlaminar toughness of composites [20,21]. Studies were found that strong fibre matrix interface bonding could improve the fracture toughness of composites, and poor adhesion strength between fibre and matrix which was caused by moisture absorption could lead to a lower fracture toughness of fibre composites [22,23].

This research is a continuation of the authors' previous work to study the PP resin modified with novel IFR consist of APP and PETAT in mechanical, thermal and flame retardant properties, now extends to the CGF/PP composites [14]. A literature and information search indicates that the effect of IFR on the mechanical and flame retardant properties of the CGF/PP composites has not yet been investigated. In this work, novel IFR (APP / PETAT) was introduced into the PP matrix to solve the "wicking actions" of CGF/PP composites. The Mode I interlaminar fracture toughness and flame retardant behaviour of CGF/PP composites modified by the IFR (CGF/PP/IFR) were studied. CGF/PP/IFR unidirectional prepregs (UD) were fabricated using an effective hot-melt impregnation method; CGF/PP/IFR laminate composites were prepared by a hot-press approach. The effects of the APP/PETAT system on the mode I interlaminar fracture toughness, and flame-retardant properties of the CGF/PP/IFR laminate composites were investigated by a double cantilever beam test (DCB), scanning electron microscopy (SEM), dynamic mechanical analysis (DMA), limiting oxygen index (LOI), vertical burning test (UL-94), and cone calorimetric test (CCT).

2. Materials and experiment

2.1. Materials

Polypropylene (KP503), a granulated product with a melt flow index of 60.0 g/10 min (230 °C, 2.16 kg), was supplied by China Petroleum and Chemical Corporation. Maleic anhydride-grafted polypropylene (MAPP), CMG9801, a granulated product with a melt flow index of 12.0 g/10 min (230 °C, 2.16 kg), was obtained from Fine-blend Compatilizer Jiangsu Co., Ltd. APP was purchased from Sinopharm Chemical Reagent Co. Ltd. PETAT was synthetised in our lab [14]. Glass fibre direct rovings (4305S), pre-treated by sizing agent containing amine group (–NH_2) and dedicated to the PP resin, were supplied by Chongqing Polycomp International Corporation.

2.2. Specimens preparation

The hot-melt impregnation process is a highly efficient and low-energy method to prepare high-quality, continuous fibre reinforced thermoplastic UD [24]. Although traditional methods have successfully prepared continuous fibre-reinforced thermoplastic composites, most consist of at least four steps [7,9]. The hot-melt impregnation process can integrate the resin modification and fibre impregnation process into one step by using an extrusion method, utilizing a twin-screw extruder

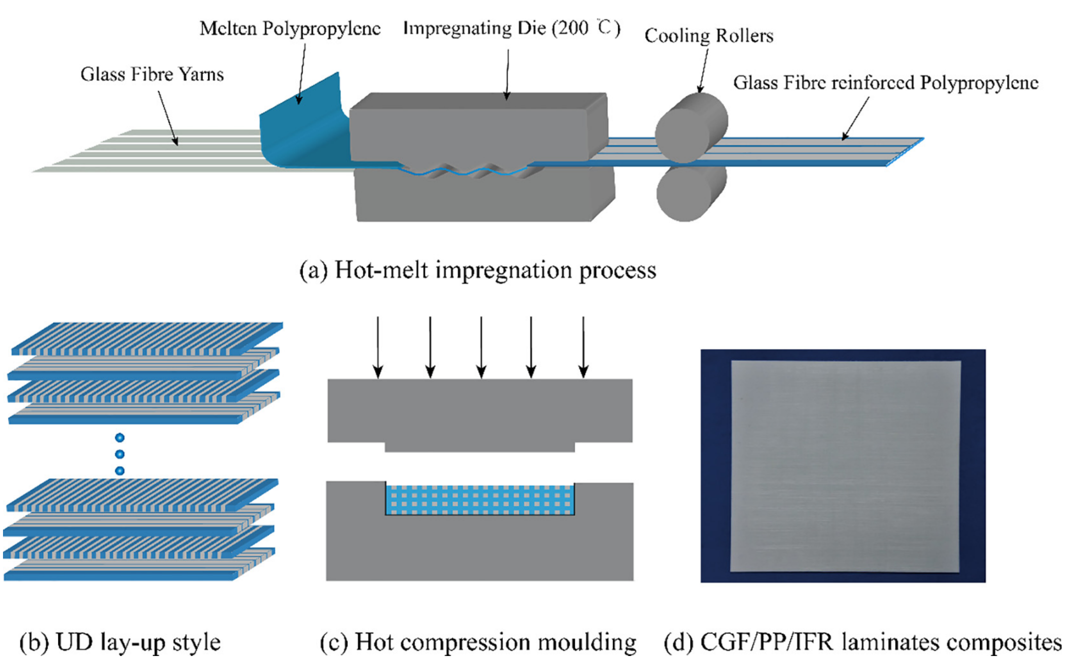

Fig. 2. Schematic illustration for the manufacture process of CGF/PP/IFR laminate composites.