

## Aging of the Flame-Retardant Properties of Polycarbonate and Polypropylene Protected by an Intumescent Coating

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**ABSTRACT:** Flame retardancy (FR) in polycarbonate (PC) and polypropylene (PP) was obtained through the application of an intumescent coating on the polymeric substrate. A better performance was obtained with PC, a char former and highly viscous polymer, compared to with PP. Indeed, whereas 61  $\mu\text{m}$  was required to obtain good FR (by the UL94 V0 rating, in particular) in the case of PC, at least 158  $\mu\text{m}$  needed to be used to give FR to PP. The aging of the coated materials induced by UV-filtered light radiation was then studied. This exposure led to a decrease in the FR. This effect was more pronounced in the case of PP compared to that of PC. The decrease in the FR was attributed to a decrease in the adhesion of the coating on the polymeric substrate because of its suspected physical aging. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 39566.

**KEYWORDS:** aging; coatings; flame retardance

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### INTRODUCTION

Since the beginning of the 20th century, there have been great developments in the field of macromolecular chemistry. Because of the possibility of developing customized polymers, natural and synthetic polymer materials have been used in a number of areas. The massive use of polymer materials in our everyday life is driven by their remarkable combination of properties, low weight, and ease of processing. Plastics are used in the packaging sector, for construction, in the electrical and motor industries, for household goods, in the agricultural and furniture industries, and other sectors.

However, an important drawback of those materials is their relatively high flammability, which is due to their chemical structure (polymers are mainly composed of carbon and hydrogen), and in most of the sectors previously discussed (e.g., construction, transport) materials have to comply with standard fire tests to be used. To reduce the combustibility of the plastics, the addition of flame retardants is usually the solution of choice.<sup>1</sup> Among them, intumescent additives can be used with various success in numerous polymers.<sup>2–4</sup> The intumescent phenomenon describes a material that, when exposed to a heat source, degrades with the subsequent formation of an expanded and porous structure that protects the underlying material.<sup>5</sup> This approach has been used in the coating industry for several decades for the protection of various substrates, including steel and

wood.<sup>6,7</sup> An intumescent coating (IntCoat) appears as a traditional paint finish, but if it is exposed to fire, it increases in volume (its thickness can increase 1000% and more; Figure 1); this leads to the formation of an expanded char at the surface of the substrate. This char exhibits a low thermal conductivity and thus limits the heat transfer from the fire to the substrate.

The use of IntCoats to protect the polymeric matrix has been poorly investigated up to this point. Only recently has an approach consisting of the use of such a coating treatment to add FR to a polymer been reported.<sup>8–12</sup> However, this approach is particularly interesting because it consists of an easy process, it does not affect the intrinsic properties of the materials (e.g., the mechanical properties), and it can be applied on a variety of polymeric matrices, including reinforced thermoplastic composites.

In this study, we investigated the FR of polymeric materials coated with an intumescent varnish and the durability of these properties when the material was exposed to UV-filtered light radiation. Polypropylene (PP) and polycarbonate (PC) were selected as two different matrices because one was a nonpolar polymer and the other one was not. Both these polymers were good examples that were representative of the limits of actual bulk treatments. PP is widely used in many fields, including as the building, automotive, electronics, and electric industries. It is highly combustible and needs to be flame-retarded. Until



**Figure 1.** IntCoat after and before exposure to fire.

recently, halogen-containing compounds, alone or in conjunction with antimony trioxide, were the main flame retardants used for PP.<sup>13</sup> Phosphorus- and nitrogen-based compounds and metal hydroxides now constitute a rapidly growing group of flame retardants and have been the focus of public interest concerning environmental friendly chemicals. Metal hydroxides,<sup>14</sup> mainly magnesium hydroxide and aluminum hydroxide, are commonly used to add flame retardancy (FR) to polymers because of their low toxicity and cost. However, usually more than a 60 wt % loading of metal hydroxides is required in PP to obtain efficient flame-retardant properties, and such high loading levels lead to a great decrease in the mechanical properties of the filled polymer materials.<sup>15–17</sup>

The same kind of observations could be made for PC. PC is known for its transparency, and it exhibits excellent mechanical strength and good electrical properties and is widely used in a variety of fields, such as in electric and electronic machinery, automobiles, and architecture. The aim to meet safety criteria in these various applications is to develop a PC-based component that is an efficient flame-retardant system and that prevents dripping during fire and allows maintenance of the mechanical and optical properties of PC. PC resin is also usually fire-retarded by the incorporation of a flame retardant during processing. The most common flame retardants used are bromine-based (e.g., decabromodiphenyl ether).<sup>18</sup> Another method is the addition of a relatively large amount of phosphorous-based flame retardants (10–30 wt %). However, these phosphorous-based additives can lower the impact strength of PC or yellow it in high-temperature or high-humidity conditions. Sodium and potassium perfluoroalkanesulfonic<sup>19</sup> acids were found to be effective in amounts well under 0.05–0.5%, but these components are still halogenated compounds. Some recent studies have described the development of fire-retardant PC combined with polymethylphenylsilsesquioxane spheres,<sup>20</sup> but the mechanical resistance issue still remains.

In this study, to validate the concept of plastic protection by an IntCoat, the results, in terms of both adhesion and fire barrier properties, of the application of a waterborne, transparent, intumescent varnish (Pyroplast HW100, Rütgers Organics, unknown formula) on flamed PP and PC samples were studied. In the first part of this article, the influence of the coating thickness on the fire-retardant properties are discussed. Then, in the second part, the aging of the materials is covered.

## EXPERIMENTAL

### Materials

The polymeric matrices used in this study were a PP (PPH7060, Total Petrochemicals) and a PC (Makrolon, Bayer). The IntCoat (Pyroplast HW100) was supplied by Rütgers Organics GmbH.

Pyroplast HW 100 is a waterborne IntCoat design for the protection of internal natural timber surfaces against the effect of flame spreading, fire propagation, and smoke emission. It is suitable for all types of softwood, hardwood, and wood derivatives. Plates of PP and PC ( $0.3 \times 10 \times 10$ ,  $0.16 \times 1.3 \times 14$ , and  $0.08 \times 10 \times 10 \text{ cm}^3$ ) were obtained by compression molding with a Daragon press (15 kN for 3 min and then 40 kN for 5 min at 195°C for PP and at 250°C for PC).

### Coating Deposit

Before applying the IntCoat on the polymer substrate, we treated the surfaces of the plates and bars with a flaming apparatus. This treatment was carried out to clean the surface of organic residues, to oxidize the surface, and thus to facilitate the adhesion of the coatings. The flaming treatment did not degrade the polymer surface; no color change or charring was observed.

An IPROS flaming apparatus was used with the following parameters: 200 mm/s, propane/air ratio = 23.5, and with two passes for PP and three passes for PC.

The coatings were then applied with a semi-industrial spraying machine from CLID. The following parameters were used: average speed = 20 rpm, jet pressure = 2 bar, pump pressure = 1 bar, cone pressure = 3 bar, product pressure = 6 bar. The spraying time was varied at values of 3, 6, 12, and 18 s to modify the thickness of the coating.

### Coating Characterization

The thickness of the coating was evaluated with a digital microscope (VHX-1000 microscope, Keyence). The coated plates were cut with a saw into small squares. To obtain a sharp edge, the borders were polished with fine sandpaper. The heights were measured with the software of the VHX-1000 microscope. The results were reproducible at  $\pm 5 \mu\text{m}$ .

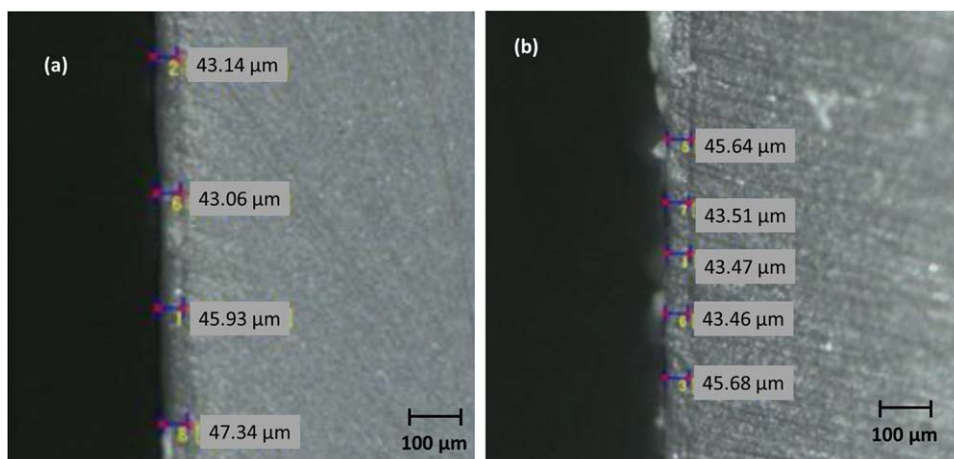
The adherence of the film on the substrate was evaluated according to ASTM D 3359-02.

### Fire Testing Methods

Limiting oxygen index (LOI; the minimum oxygen concentration needed to support the candlelike combustion of plastics) was measured with a Fire Testing Technology instrument on sheets ( $100 \times 10 \times 3 \text{ mm}^3$ ) according to the standard oxygen index test (ISO4589).

The UL-94 classification was obtained on sheets ( $100 \times 12.7 \times 1.6$  or  $0.8 \text{ mm}^3$ ) according to the conditions of a standard test (ASTM D 3801), that is, in a vertical position (the bottom of the sample was ignited with a burner). This test provided only a qualitative classification of the samples (V-0, V-1, and V-2).

A Fire Testing Technology mass loss calorimeter was used to carry out measurements on the FR samples according to the procedure defined in ASTM E 906. The equipment was identical to that used in oxygen consumption cone calorimetry (ASTM E 1354-90) except that we used a thermopile in the chimney to obtain the heat release rate (HRR) rather than using the oxygen consumption principle. Our procedure involved exposure of specimens (coated face) in a horizontal orientation. An external heat flux of  $50 \text{ kW/m}^2$  was used to run the experiments. This



**Figure 2.** Optical picture of the cross section of the coated (a) PC and (b) PP for a spraying time of 3 s. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

flux corresponded to the common heat flux in a developed fire scenario. A distance of 55 mm was used to allow the expansion of the IntCoat. The mass loss calorimeter was used to determine HRR. When measured at  $50 \text{ kW/m}^2$ , the HRR was reproducible to within  $\pm 10\%$ . The data reported in this article are the averages of three replicated experiments.

#### Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectra were recorded with a Nicolet 400D spectrometer in the range  $4000\text{--}300 \text{ cm}^{-1}$  in an attenuated total reflectance mode. The IR spectra were then plotted and analyzed with the OMNIC program.

#### Thermogravimetric Analysis (TGA)

TGA was performed with a TA Instruments Q 5000 IR microbalance. Samples of 10 mg were heated in open silica pans under a nitrogen flow at heating rate of  $10 \text{ K/min}$  from room temperature to  $800^\circ\text{C}$ . The parameters obtained from the TGA were the remaining mass as a function of temperature, the mass loss rate as a function of temperature, the maximal mass loss rate temperature, and the residual yield at  $800^\circ\text{C}$ .

#### Aging

The aging of the coated materials was evaluated with a Q-SUN xenon test chamber from Q-LAB. (UV filter for 200 and 400 h). The bars for the UL94 measurements after the aging treatment were turned every 100 h to get the same light radiation for both sides of the bars. The bars were tested after 200 and 400 h of exposure.

## RESULTS AND DISCUSSION

### Effect of the Thickness of the Coating on the Flame-Retardant Properties of the Materials

To investigate the effect of the thickness of the coating and to determine the optimized thickness that permitted the achievement of a satisfactory fire performance, the spraying time was varied during the deposition. The thickness of the coating was evaluated with an optical microscope, and Figure 2 presents, as an example, the results obtained for a spraying time of 3 s.

We observed that whatever the matrix, the thickness of the coating was homogeneous along the sample. It varied from 41

to  $47 \mu\text{m}$  on PC and from 41 to  $45 \mu\text{m}$  on PP. Several observations were done, and a classical statistical calculation showed that for a spraying time of 3 s, the thickness was equal to  $42 \pm 5 \mu\text{m}$ .

The evaluation of the thickness versus spraying time is reported in Table I. The variation of the coating thickness on the plate could be considered equal to  $\pm 5 \mu\text{m}$ .

Figure 3 reports the LOI of PP and PC versus the coating thickness. The LOI is the minimum concentration of oxygen, expressed as a percentage that supports the combustion of a polymer: the higher the LOI value was, the better the FR of the sample was.

We observed that higher values were obtained for PC compared to PP in both cases (virgin and coated materials). This difference made sense because the behavior of both materials against fire is very different. Whereas PC chars when it is exposed to a flame, PP melts and burns vigorously after ignition. Lyon et al.<sup>21</sup> compared the flammability of various polymers, including PP and PC, using different techniques. They found the char yields of PP and PC with a microscale calorimeter, and they reported values of 0 and 23 wt %, respectively. On the other hand, the total heat released by combustion of a volatile fuel from PP was reported to be equal to around  $43 \text{ kJ/g}$ ; in contrast, that from PC was around  $20 \text{ kJ/g}$ . This demonstrated that the flammability of PC and PP were different, and thus it was not surprising to observe differences in their FR.

**Table I.** Thickness of the Coating versus the Spraying Time

Spraying time (s)	Coating thickness on PC ( $\mu\text{m}$ )	Coating thickness on PP ( $\mu\text{m}$ )
3	42	42
6	61	64
12	131	137
18	157	158

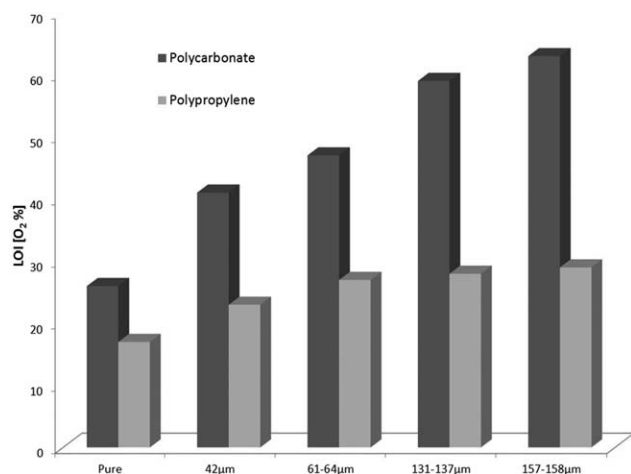


Figure 3. LOI versus coating thickness for PC and PP.

The LOI value of virgin PC was 26 vol %, and it increased to 41 vol % for PC with a 42- $\mu\text{m}$  coating and up to 63 vol % for PC with a 157- $\mu\text{m}$  coating. The LOI of virgin PP was 17 vol %, and it increased from 23 up to 29 vol % when the thickness increased from 42 to 158  $\mu\text{m}$ , respectively. In both cases, an improvement in the FR of the materials was observed with increasing coating thickness, and this improvement was higher in the case of PC (with LOI increases of 22 vol % for a 157  $\mu\text{m}$  thick coating compared to virgin PC) than for PP (with an LOI gain of 12 vol % for a 158  $\mu\text{m}$  thick coating compared to virgin PP). Those results demonstrate that the use of an IntCoat to improve the FR of PP and PC was efficient.

Table II reports the UL94 rating for both the 0.8 and 1.6 mm thick samples. UL94 is a plastic flammability standard widely required in the field of electric and electronic equipment. The rating varies from V0 for a low flammable material (in which burning stops within 10 s on a vertical specimen; drips of particles are allowed as long as they are not inflamed) to nonclassified (NC) material (in which the burning time is higher than 30 s on a vertical specimen and/or the material burns completely). To achieve a V0 rating in PC or PP, various additives (e.g., halogen-, phosphorus-, or silicon-based additives) can be added in the bulk materials. However, large quantities (5–30 wt

Table II. UL94 Rating versus the Coating Thickness for PC and PP

Substrate	Coating thickness ( $\mu\text{m}$ )	UL94 rating (0.8-mm bars)	UL94 rating (1.6-mm bars)
PC	0	V2	V2
	42	V0	V0
	61	V0	V0
	131	V0	V0
	157	V0	V0
PP	0	NC	NC
	42	V2	NC
	64	V2	V2
	137	V2	V1
	158	V1	V0

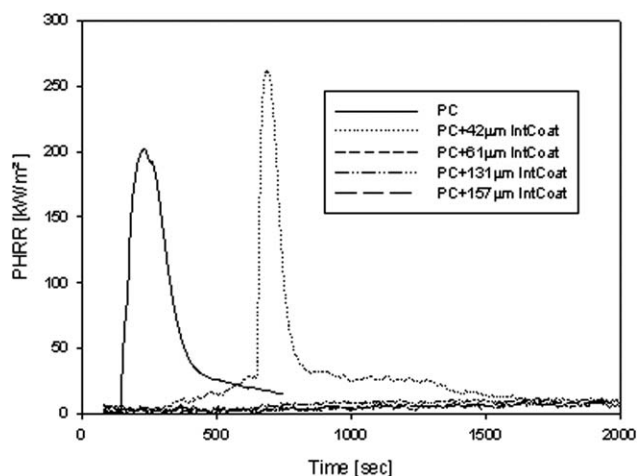
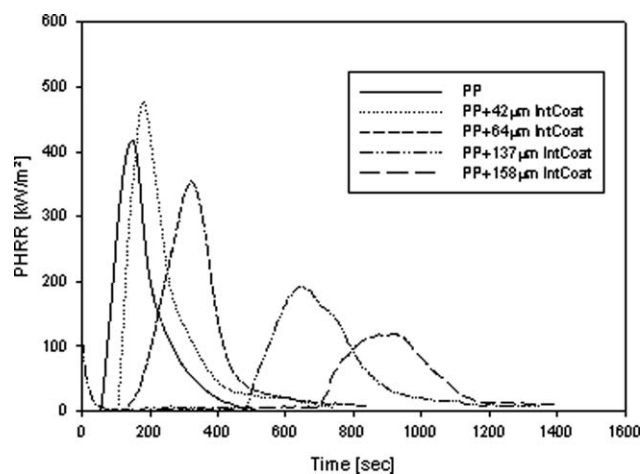


Figure 4. HRR versus time for the PC and coated PC versus the coating thickness.

%) are often required to achieve such performance, and these quantities affect many of the desirable physical and mechanical properties of the matrix.<sup>22,23</sup> As observed in the case of LOI, better properties were obtained in the case of PC compared to that of PP. Indeed, whereas for PC, V0 was achieved for both the 0.8 and 1.6 mm thick samples whatever the thickness of the IntCoat, V0 was only achieved in the case of PP for the 1.6-mm bar and the 158  $\mu\text{m}$  thick IntCoat. Similarly to what was discussed previously, this difference could be explained by the burning behavior of the virgin matrix. Indeed, because PC is a highly viscous and charring material, when the material was exposed to the flame, a low deformation of it occurred, and thus, the intumescent char layer could act as a protective coating. In the case of PP at the opposite, when the material degraded, its viscosity sharply decreased, and drops were observed. In that case, the development of the IntCoat was not fast enough to prevent the increase in temperature of the substrate and thus the melting of the PP. The specimen was highly deformed, and the intumescent char could not totally follow this deformation. As a result, mass and heat transfers could occur between the flame and the substrate, and this resulted in a low FR. However, the achievement of a V0 rating in the case of PP was not an easy task, and thus the results obtained for the 158  $\mu\text{m}$  thick IntCoat were very encouraging.

Finally, a mass loss calorimeter was also used to evaluate the FR of the coated PC and PP. HRR curves as a function of the time are reported for PC and PP in Figures 4 and 5, respectively, and corresponding data are summarized in Table III. HRR and its maximum value [peak heat release rate (PHRR)] can be used to characterize the hazard represented by a material with regard its contribution to fire and can provide information on the fire size and fire growth rate: the lower the HRR is, the better the fire performance is.

Similar to the results obtained for the two other fire testing methods, the FR of the coated materials were better in the case of PC than in that of PP. In particular, we observed that for PC, when a thickness of 61  $\mu\text{m}$  or higher was used, no ignition of the material occurred, and thus, the HRR remained close to



**Figure 5.** HRR versus time for the PC and coated PC versus the coating thickness.

zero. For a thickness of 42  $\mu\text{m}$ , a peak was observed with a slightly higher PHRR value compared to that of virgin PC (260 vs 200  $\text{kW/m}^2$ ), but PHRR was observed at a longer time (720 vs 230 s). This correlated well with the time to ignition (TTI), which was around 140 s for virgin PC and around 650 s for PC coated with 42  $\mu\text{m}$  of intumescent varnish. We could thus assume that when the plate was coated with 42  $\mu\text{m}$  of intumescent varnish, the intumescence developed; this led to the formation of a protective layer, but this layer was too thin and fragile. Just before ignition, the protective layer broke because of internal pressure, and this led to the release of the degradation products of PC that were previously trapped by the IntCoat and ignition occurred. As a result, this led to the rapid ignition of these gases, and their combustion led to a slightly higher PHRR.

In the case of PP, the results were very different because, in all cases, ignition was observed, and a peak corresponding to the combustion of the material occurred. The TTI increased with the coating thickness, whereas the PHRR decreased. Indeed, pure PP ignited after 56 s and reached a maximum HRR of 420  $\text{kW/m}^2$ . The 42  $\mu\text{m}$  thick coated PP ignited after 107 s and reached an HRR value of 477  $\text{kW/m}^2$ . This value was a bit higher than for pure PP, and a similar explanation to that in

**Table III.** MLC Data of the PC- and PP-Coated Materials

Substrate	Coating thickness ( $\mu\text{m}$ )	PHRR ( $\text{kW/m}^2$ )	TTI (s)
PC	0	200	140
	42	260	650
	61	—	—
	131	—	—
	157	—	—
PP	0	420	56
	42	477	107
	64	355	137
	137	188	506
	158	118	705

the case of PC was proposed. For the 64  $\mu\text{m}$  thick coating, PHRR was slightly lower than for the virgin PP (355  $\text{kW/m}^2$ ). For the 137- and 158- $\mu\text{m}$  coating thicknesses, the PHRR values were 188 and 118  $\text{kW/m}^2$ , respectively.

From these results, we also concluded that a minimum thickness of 158  $\mu\text{m}$  was required to achieve satisfactory results (UL94 V0 rating, high LOI, low PHRR value, and high TTI) in the case of PP, whereas 61  $\mu\text{m}$  was the minimum thickness for PC. PP and PC behave in completely different ways when exposed to fire. Indeed, it was mentioned previously that PP melts before burning, whereas PC, which is a highly viscous material, does not (only softening is observed). Thus, the cohesion between the protective layer and the polymer during the mass loss calorimetry (MLC) test was lower. As a result, degradation gases (fuel) could escape, and this could lead to the ignition of the material.

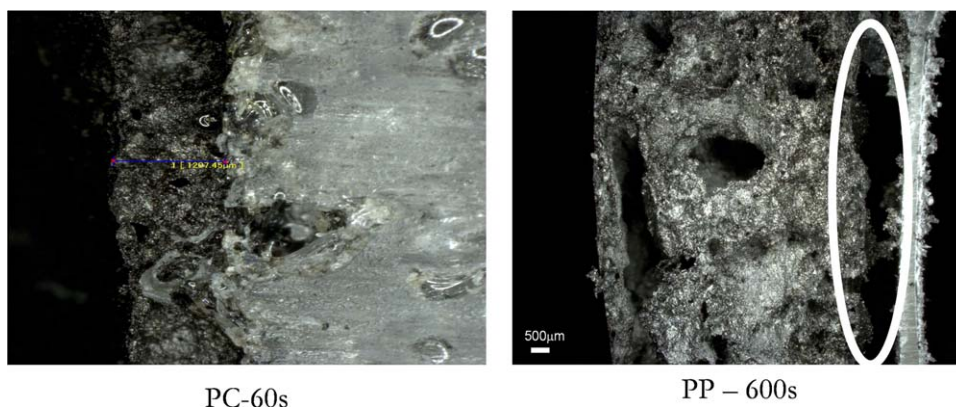
To confirm the protective mechanism of the IntCoat and to better understand the role of the polymer matrix, MLC experiments were stopped after the characteristic time of combustion. The sample was thus taken out from the heat source (conical heater) and quickly covered to cut off the oxygen supply and lead to fast flame suppression. The collected residues were analyzed with a digital optical microscope, and the results are presented in Figure 6. As shown in these pictures, in the case of PC, the intumescent paint stuck to the polymer matrix and thus could play its protective role. When we observed the sample after a longer combustion time (results not shown), the only difference was in the thickness of the IntCoat. The residue collected at the end of the MLC test still showed some unburned PC. At the opposite, in PP, just before ignition (at time = 600 s), a gap was observed between the intumescent paint and the polymer matrix. We observed that the big bubble between the matrix and the coating (shown as a white ellipse in the picture) increased in size up to the break of the IntCoat, which led to the release of fuel and thus to the ignition of the material. At the end of the test, no remaining virgin PP was observed.

### Aging

After the optimization of the coating thickness, the aging of the coated PP and PC was evaluated. Materials coated with 61  $\mu\text{m}$  for PC and 158  $\mu\text{m}$  for PP were chosen for this part of the study. The aging conditions were fixed to simulate the aging inside a building (with a UV filter for 200 and 400 h). After these treatments, the fire-retardant properties of the coated samples were evaluated with MLC and UL94. Additionally an adhesion test was performed to determine the influence of aging on the adhesion between the paint and the substrate. A film of pure intumescent varnish was also exposed to aging under the same conditions to study the stability of the varnish under UV-filtered light. The aged film was characterized with FTIR spectroscopy.

**FR of the Aged Materials.** Table IV reports the UL94 rating of coated PP and PC after 200 and 400 h of exposure to UV-filtered light.

We observed that whereas the FR of the PC-based materials were not affected by the aging, this was not the case for the PP-



**Figure 6.** Digital pictures of the cross section of the 61- $\mu\text{m}$  coated PC and 158- $\mu\text{m}$  coated PP 60 and 600 s, respectively, of exposure with MLC (50  $\text{kW}/\text{m}^2$ ). [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

based materials. Indeed, when the coated PP was exposed for 400 h to UV-filtered light, it achieved a V1 rating (without any dripping) and a V0 rating at the initial stage and after 200 h of exposure.

HRR curves of the coated PC and PP before and after 200 h of exposure to UV-filtered light are reported in Figures 7 and 8, respectively. These curves demonstrate that for both PC and PP, the FR of materials was affected by the aging treatment.

In the case of PC, whereas no ignition was observed for the coated PC, as previously discussed, the ignition of the material occurred for the aged material around 2200 s (>30 min). So, even when the FR of the coated PC was very good, this demonstrated that the UV-filtered light aging could affect the properties at long times.

In the case of PP, similar results were observed. Indeed, the TTI decreased from 705 s for nonaged material to 164 s for aged material. The value of the PHRR was, however, only slightly affected by the aging treatment (134  $\text{kW}/\text{m}^2$  for the aged material vs 118  $\text{kW}/\text{m}^2$ ).

Those results demonstrate that the FR of the intumescent coated materials was affected by the exposure to UV-filtered light. To better understand this effect, at least partially, adhesion tests of the coating after aging were first carried out.

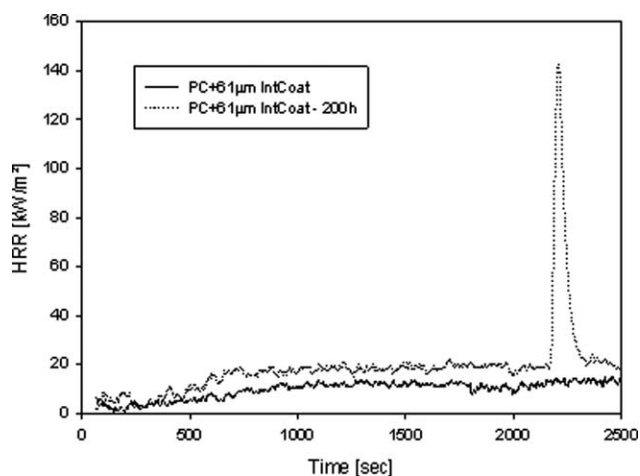
**Adhesion of the Intumescent Varnish versus Aging.** The adhesion of the coating was evaluated on PC and PP before and

**Table IV.** UL94 Rating of the Aged Materials

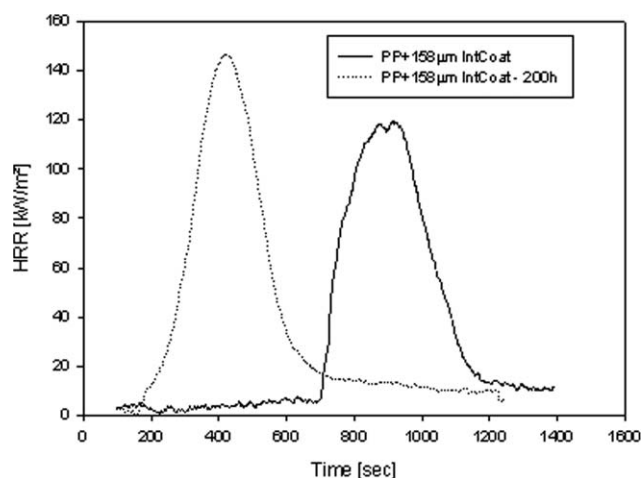
Material	Aging time (h)	UL94 rating (1.6 mm bars)
PC + 61- $\mu\text{m}$ IntCoat	0	V0
	200	V0
	400	V0
PP + 158- $\mu\text{m}$ IntCoat	0	V0
	200	V0
	400	V1

after aging with the tape test. Pictures of the materials and their classification obtained according to ASTM D 3359-02 are reported in Table V.

The adhesion of the varnish on PC achieved the best classification (5B); this demonstrated a good adhesion between the coating and the substrate. After 200 h of UV-filtered light exposure, the picture of the coated PC plate showed a clear difference with the virgin sample. The adhesion decreased after aging, and the specimen was classified 3B. After 400 h of UV-filtered light treatment, the adhesion did not change significantly compared to that of the sample treated for 200 h (the sample was still classified 3B). These results thus demonstrate that the adhesion of the varnish decreased after UV-filtered light exposure. Similar results were obtained in the case of PP. Indeed, the adhesion test of the virgin coated PP showed slightly worse adhesion compared to PC because the sample was classified 4B. However, this result was sufficient to obtain good FR, as demonstrated in the previous section. The adhesion of the PP sample after 200 h was affected by the UV-filtered light treatment, and the sample only reached a 2B rating. This aging of the adhesion was even more pronounced after 400 h because, in that case, a very low



**Figure 7.** HRR versus time for the coated PC before and after 200 h of exposure to UV-filtered light.



**Figure 8.** HRR versus time for the coated PP before and after 200 h of exposure to UV-filtered light.

adhesion of the varnish was obtained, and the material was classified 1B.

We thus had evidence that the adhesion was affected by the UV-filtered light exposure and that this exposure resulted in a decrease in the FR of the materials. Moreover, some previous studies have demonstrated that adhesion is a key parameter in the FR of a material with IntCoats.<sup>8,10</sup>

**Characterization of the Aging of the Intumescent Varnish.** To evaluate whether the decrease in FR should also have been attributed to a chemical modification of the intumescent varnish, its aging was studied through analysis of the FTIR spectra of both nonaged and aged coatings (Figure 9) and its thermal stability according to TGA (Figure 10).

The IntCoat used in this study was a commercial product. Indeed, the objective of the study was not to develop a new IntCoat but to validate the use of such an efficient coating in the field of polymer FR. Thus, it was not possible to attribute all of the FTIR peaks of the spectra, but some conclusions, however, could be drawn. In particular, we observed that the spectra for

the nonaged and aged coatings were very similar. However, we observed that with UV-filtered light exposure, three peaks appeared at 1976, 2027, and 2158  $\text{cm}^{-1}$ . These three peaks were attributed to the stretching vibrations of triple bonds ( $\text{C}\equiv\text{C}$ ,  $\text{C}\equiv\text{N}$ ) and/or of cumulated double bonds ( $x=y=z$ ). However, because we did not have any information on the composition of the varnish, it was not possible to explain the formation of such species further.

TGA (Figure 10) was also carried out to study the aging of the intumescent varnish.

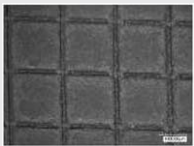


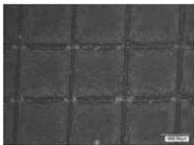


Similar to what we discussed previously, it was not possible to clearly determine the degradation scheme, but the objective was to see whether the TGA curves were very different before and after aging. Indeed, if the aging of the FR was due to chemical aging of the intumescent paint (e.g., loss of the active ingredient), we should have noticed a clear difference in the TGA curves. This was not the case for the intumescent varnish aged for 200 or 400 h. Indeed, in both cases, the thermal stability was similar.

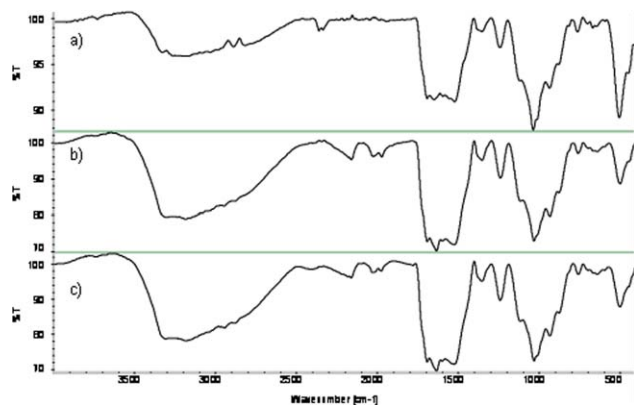
### Discussion

It was thus demonstrated that good FR could be achieved through the use of an IntCoat on a PC or PP matrix. The intumescent structures developed in case of fire acted as a protective barrier. Better performance could be achieved for PC compared to PP. Indeed, whereas 61  $\mu\text{m}$  of intumescent varnish allowed us to achieve good FR in the case of PC, at least 158  $\mu\text{m}$  was needed in the case of PP. This was attributed to differences in the fire behavior of the two materials. Indeed, whereas PP melted and burned vigorously, PC charred when it was degraded. As a result, the protective intumescent layer could be maintained in the case of PC whereas for PP, the high pressure due to gas release and the deformation due to melting led to its degradation after some time (depending on the coating thickness).

The effect of UV-filtered light exposure was analyzed in a second step. It was demonstrated that the aging led to a decrease in the FR of the material. This loss of efficiency was attributed to a decrease in the adhesion properties of the coating versus aging rather than to a chemical aging of the intumescent varnish. There

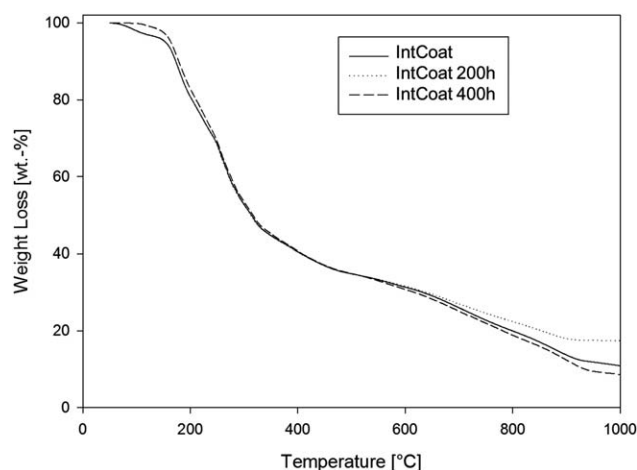
**Table V.** Adhesion Properties of the Intumescent Varnish on PC and PP versus the Aging Time

Material	Virgin material	After 200 h of exposure	After 400 h of exposure
PC	 5B	 3B	 3B
PP	 4B	 2B	 1B



**Figure 9.** FTIR spectra of the intumescent varnish (a) nonaged, (b) after 200 h of UV-filtered light exposure, and (c) after 400 h of UV-filtered light exposure. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

have been a number of studies reported in the literature on the chemical aging of coatings caused by different types of radiation with coatings of various natures (e.g., epoxy, acrylic) and compositions. It has generally been observed that the modification of the chemistry of the coating due to aging leads to some modification of the coating properties that negatively affect the adhesion. With IntCoats, even though only few studies have been reported on the aging of such coatings,<sup>24–27</sup> it is generally proposed that the hydrophilic components of the IntCoats move to the surface of the coating and can be dissolved in the environmental media (e.g., water, air); this leads to a decrease in the FR of the coatings. This should not have been the case in our study because the aged material exhibited similar expansion compared to the nonaged material; this demonstrated that the intumescent process occurred in a similar way. Moreover, according to the FTIR analyses carried out in this study, it appeared that under our aging conditions (UV-filtered light radiation), the chemistry of the coating was only slightly affected by the aging treatment. Thus, physical aging (the relaxation process of the resin used in the intumescent varnish) was suspected. Indeed, it was reported that when any organic coating is subjected to physical aging, that



**Figure 10.** TGAs of the intumescent varnish nonaged, after 200 h of UV-filtered light exposure, and after 400 h of UV-filtered light exposure.

aging will further affect the coating properties (e.g., rigidity, fracture resistance).<sup>28</sup>

## CONCLUSIONS

- The use of IntCoat to flame-retard thermoplastic polymers is a promising and efficient approach that should be further developed.
- The properties obtained depended on the nature of the polymeric matrix.
- Better performance could be obtained with PC compared to PP.
- FR decreased when the materials were exposed to UV-filtered light radiation.
- The decrease in FR was attributed to a decrease in the adhesion of the varnish rather than to its chemical modification.

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