

# Application of Chlorinated Waste Rubber as a Flame Retardant of Low Density Polyethylene

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**ABSTRACT:** For the reuse of the abundant waste rubber (WR), chlorinated waste rubber (Cl-WR) was prepared by a water-based chlorination method using chlorine gas as the chlorinating reagent. The resultant Cl-WR was used as a flame retardant of low density polyethylene (LDPE) matrix. The properties of WR and Cl-WR were characterized by thermogravimetric (TG) and Fourier transform infrared (FTIR) spectrometer. The results indicated that the Cl-WR with a 42% chlorine content had a higher char residue than WR. The limiting oxygen index (LOI) value can be increased up to 27.2 and the flame retardancy reached to the maximum (V-0) grade for the LDPE/Cl-WR-Sb blends

with mass ratios of Cl-WR/LDPE and Cl-WR/Sb<sub>2</sub>O<sub>3</sub> being 80/100 and 1.5/1, respectively. The improvement of the flame retardancy and thermal properties of the LDPE/Cl-WR-Sb blends were attributed to the char formation and synergistic effect between Cl-WR and Sb<sub>2</sub>O<sub>3</sub> in a high temperature region, although the mechanical properties of the said blend decreased in comparison with that of the neat LDPE. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 123: 3495–3502, 2012

**Key words:** waste rubber; flame retardant; chlorinated waste rubber; synergistic effect

## INTRODUCTION

For the sustainable development of modern society and persistent protection of future environment, the mission of the waste rubber (WR) disposal is becoming more and more urgent all over the world and thus serious attention has been attracted in recent years.<sup>1–4</sup> In retrospect, pyrolysis and incineration techniques<sup>5–7</sup> and polymer blend technology<sup>8,9</sup> are the two most important approaches for cutting down the accumulation of WR. As far as the former is concerned, the available components of WR are involved in energy,<sup>10,11</sup> carbon black,<sup>12–14</sup> and metal.<sup>6,15</sup> However, a major drawback of this process is the concomitant releases of unmanageable masses of sulfur oxides (SO<sub>x</sub>), nitrogen oxides (NO<sub>x</sub>), and other polycyclic aromatic hydrocarbons. These emissions demand further treatment to conform to the strict environmental regulations. Therefore, in view of the economic and environmental advantages, the

polymer blend technology is a better choice. A number of thermoplastics/WR blends have been successfully prepared with some improving properties. Mészáros et al. investigated the effect of ethylene vinyl acetate copolymer (EVA) on the processing parameters and the mechanical properties of low density polyethylene (LDPE)/ground tire rubber (GRT) blend.<sup>16</sup> Similarly, Kim et al. investigated the mechanical and dynamic mechanical properties of a WR powder-filled high-density polyethylene (HDPE) composite.<sup>17</sup>

Surface modification of WR has received great attention in recent years and a wide variety of modified techniques have been reported.<sup>18–21</sup> Shanmugharaj et al. conducted a research work on the surface modification of waste rubber powder surface using UV radiation and characterized the properties of polypropylene/waste powder composites. Naskar et al. studied maleic anhydride treatment of WR to prepare a composite based on ethylenepropylene-diene terpolymer (EPDM) rubber, maleic anhydride-grafted WR, and HDPE.<sup>21</sup> Furthermore, chlorination is an alternative modification method offering multifunctional properties to WR. Oldfield et al. studied chlorination of vulcanized rubber surface using trichloroisocyanuric acid (TCICA).<sup>22</sup> Naskar et al. also employed TCICA as the chlorinating agent to modify WR surface and discovered that the modified

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TABLE I  
Recipes of the LDPE/Cl-WR-Sb Blends

Ingredients	LDPE	Cl-WR	Sb <sub>2</sub> O <sub>3</sub>	Montmorillonite	Stearic acid	Silicane coupling agent
Contents (phr <sup>a</sup> )	100	80 in total		3	3	0.8

<sup>a</sup> phr is the abbreviation of weight parts per 100 weight parts LDPE.

WR/PVC blends have better mechanical properties and dielectric constant compared with the starting speices.<sup>23,24</sup> However, these modifications are limited to the surface of WR only and little improvements are reported to the inner properties resulting in unsatisfactory properties to some extent.<sup>24</sup>

In this article, we report a modification method of WR via a chlorination process using chlorine gas as the chlorinating reagent. The modified WR, i.e., chlorinated waste rubber (Cl-WR) was used as a novel flame retardant of LDPE, a well-known flammable polyolefin. To evaluate the flame retardancy and thermal properties of the neat LDPE and its blends, Cl-WR, antimony trioxide (Sb<sub>2</sub>O<sub>3</sub>), and other ingredients (LDPE/Cl-WR-Sb blends), the limiting oxygen index (LOI), UL-94 vertical burning (UL-94) tests, and thermogravimetry (TG) are employed.

## EXPERIMENTAL

### Materials

All materials and reagents were obtained from commercial sources in China and used as received. WR with 200–250 μm particles size was kindly supplied by Tianjin Tianyuanheng Rubber Manufacturer. Industrial grade chlorine gas was obtained from Beijing Second Chemical Factory. Initiator azobisisobutyronitrile (AIBN) was supplied by Tianjin Damao Chemical Reagent Factory. Commercial LDPE (18D) pellet was obtained from Daqing Petrochemical Company, PetroChina. Antimony trioxide and

stearic acid of CP grade were purchased from Tianjin Damao Chemical Reagent Factory. CP grade silicane coupling agent was supplied by Tianjin Fuchen Chemical Reagent Factory. Montmorillonite (MMT) of CP grade was purchased from Shanxi Institute of Mine.

### Chlorination of WR

50 g of WR particles were dispersed in 300 mL water in a four-necked flask (1000 mL) equipped with a mechanical stirrer, thermometer and condenser at 70°C. After WR was well dispersed, 0.5 g of AIBN was added to the flask and chlorine gas was introduced at a fixed flow rate at atmospheric pressure until the chlorine level of Cl-WR reached equilibrium. Nitrogen gas was then introduced to strip residual chlorine gas from the reactor and pipeline. Cl-WR granules were filtered, washed thoroughly with tap water, and dried overnight at 80°C in an oven.

### Mixing and Molding

LDPE/Cl-WR-Sb blends were prepared successfully according to the literature procedure.<sup>23</sup> Totally, 100 g of LDPE and the additives (synergistic agent, lubricant, coupling agent) were mixed in proportion as shown in Table I. Melt mixing was performed at 120°C on a two-roll mill for 5 min followed by the introduction of Cl-WR granules containing 42%

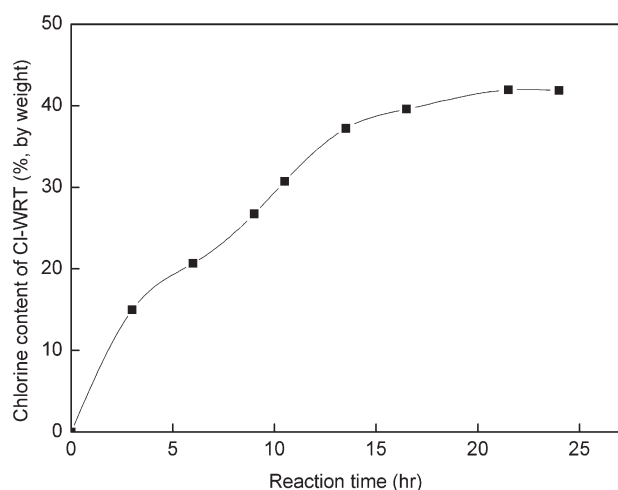


Figure 1 Chlorine content of Cl-WR versus reaction time.

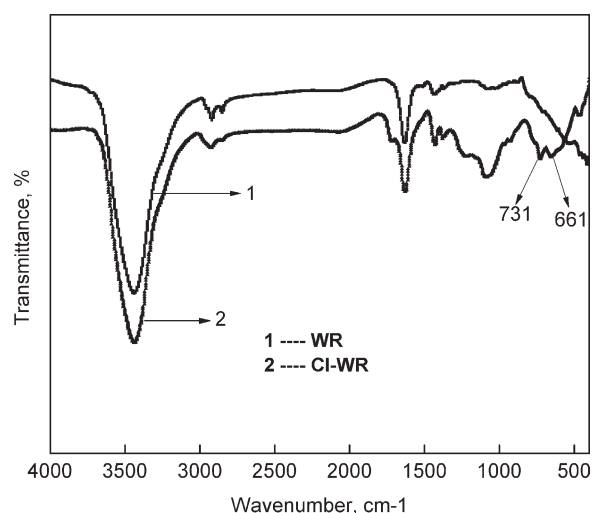


Figure 2 Infrared spectra of WR and Cl-WR.

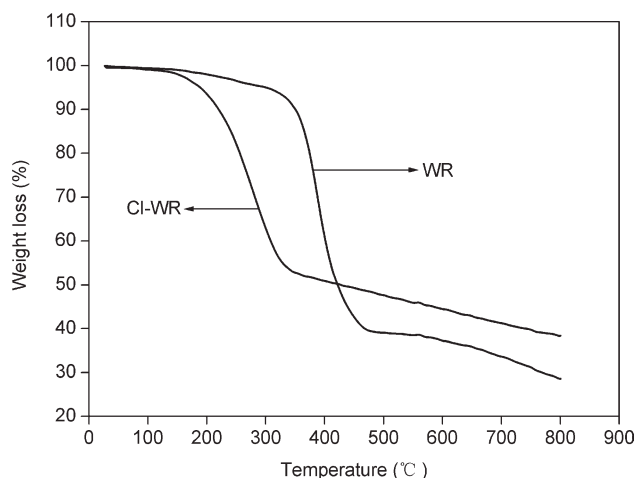


Figure 3 TG curves of WR and CI-WR.

chlorine. The sheeted out compounds were compressed and molded in an electrically heated hydraulic press at 130°C for 4 min at a pressure of 10 MPa. Cooling of the sample was performed under pressure by water circulation through the hot platens with dimensions of 180 × 120 × 3 mm<sup>3</sup>.

### Characterization

IR spectroscopy was applied with a Fourier transformation infrared (FTIR) spectrometer (Nexus 8700, USA) using potassium bromide (KBr) pellets technique. Sample and KBr were dried and mixed followed by pressing them into thin disc under hydraulic press and thus were used as a sample for FTIR measurements. The spectrum was obtained at mid-infrared region.

X-ray diffraction (XRD) analysis was carried out to comment the interlayer space of MMT in the LDPE/CI-WR-Sb blends with a X-ray diffractometer (D/Max2500, VB2+/PC, Rigaku, Japan) using Cu, K $\alpha$  radiation ( $\lambda = 0.154$  nm, 40 kV, 50 mA). Diffraction patterns were collected in reflection-mode geometry from 0.5° to 10° 2 $\theta$  at a scanning rate of 1° 2 $\theta$ /min.

Scanning electron microscopy (SEM, S-4700, Hitachi, Japan) and transmission electron microscopy (TEM, Jeol 4000, Japan) were used to characterize the dispersion of the additives and the morphology of samples.

TABLE II  
Experimental Data of the TG Scans of WR and CI-WR

Weight loss	Temperature (°C)	
	WR	CI-WR
$T_{2\%}$	201	151
$T_{10\%}$	350	220
$T_{30\%}$	388	282
$T_{50\%}$	422	428
$T_{70\%}$	770	---
Char residue (%)	28	38

TG was carried out using a TG analyzer (TG 209, NETZSCH, Germany) in the temperature ranging from room temperature to 800°C at a linear heating rate of 10°C/min and with a nitrogen flow of 50 mL/min. Samples were measured in a sealed aluminum pan with a mass of about 4 mg.

LOI corresponds to the minimum percentage of oxygen needed for combustion of the specimens in an oxygen-nitrogen atmosphere. Accordingly, LOI values were measured on a JF-3 oxygen index meter (Jiangning, China) with sheet dimensions of 100 × 6.5 × 3 mm<sup>3</sup> in accordance with ISO 4589 standard.

American National Standard UL-94 vertical burning test is commonly used to determine the ignition resistance of materials. The test results are classified by the flammability ratings of V-0, V-1, V-2, or fail (no rating). The V-0 rating indicates a material with the best flame retardancy. UL-94 tests were conducted on a vertical burning test instrument (CZF-3-type, Jiangning, China) with sheet dimensions of 125 × 12.5 × 3 mm<sup>3</sup> according to UL-94.

Dumbbell-shaped tensile test pieces were punched from the molded sheets. Tensile strength was measured according to ISO 37, by employing a Universal Testing Machine (LD30 K, LLOYD, England) at a strain rate of 500 mm/min at room temperature. In regard to the tensile strength, five test pieces were used for each sample.

## RESULTS AND DISCUSSION

### Relationship between chlorine content of CI-WR and reaction time

The flame retardancy of LDPE/CI-WR-Sb is greatly affected by the chlorine content of CI-WR and have

TABLE III  
LOI and UL-94 Data of Neat LDPE and LDPE/CI-WR-Sb Blends at Different Composition

Sample No.	Total amount of flame retardants (phr <sup>a</sup> )	Mass ratio of CI-WR to Sb <sub>2</sub> O <sub>3</sub>	LOI (%)	UL-94 test				
				$t_1$ (s)	$t_2$ (s)	$t_{1+2}$ (s)	Dripping	UL-94 rating
LDPE	0	—	17.4	>30	>30	>30	Yes	No rating
A	80	1:1	26.6	1.9	1.5	3.4	Yes	No rating
B	80	1.5:1	27.2	1.5	1.4	2.9	No	V-0
C	80	2:1	27.0	1.8	1.9	3.7	Yes	No rating

<sup>a</sup> phr is the abbreviation of weight parts per 100 weight parts LDPE.

**TABLE IV**  
**LOI Value and UL-94 Data of Neat LDPE and LDPE/CI-WR-Sb Blends at Different Loading of Flame Retardant**

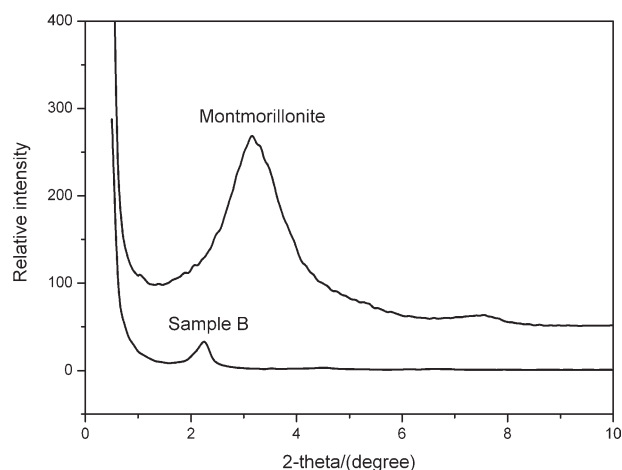
Sample No.	Amounts of flame retardant (phr <sup>a</sup> )	Mass ratio of CI-WR to Sb <sub>2</sub> O <sub>3</sub>	LOI (%)	UL-94 test				
				t <sub>1</sub> (s)	t <sub>2</sub> (s)	t <sub>1+2</sub> (s)	Dripping	UL-94 rating
LDPE	0	–	17.4	>30	>30	>30	Yes	No rating
D	50	1.5:1	24.9	22.9	>30	>30	Yes	No rating
E	60	1.5:1	25.6	10.9	>30	>30	Yes	No rating
F	70	1.5:1	26.8	5.2	20.5	25.7	No	V-1
B	80	1.5:1	27.2	1.5	1.4	2.9	No	V-0
G	90	1.5:1	27.8	1.2	1.4	2.6	No	V-0
H	100	1.5:1	28.3	1.3	1.2	2.5	No	V-0

<sup>a</sup> phr is the abbreviation of weight parts per 100 weight parts LDPE.

a direct relation between them i.e., the flame retardancy increases with the increase of chlorine content and *vice versa*. Accordingly, a series of CI-WRs with different chlorine content were prepared based on the method discussed in Experimental section at a temperature of 70°C. The relationship between chlorine content of CI-WR and reaction time was investigated and the data are shown in Figure 1. It is observed that the chlorine content of CI-WR increased steadily with reaction time at the first chlorination process which leveled off at 42% content (by weight), indicating the equilibrium of chlorination reaction under the specified conditions. Thus it is assumed that 42% is the maximum achievable chlorine content of CI-WR. Based on this fact, as a flame retardant to blend with LDPE matrix, this chlorine content was used throughout the following experiments with a reaction time of 22 h.

### Characterization of CI-WR

The microstructure of WR and CI-WR were characterized by IR spectra, as shown in Figure 2. It is seen that these IR spectra were virtually same in the wavenumber of (4000–1500 cm<sup>-1</sup>), while they



**Figure 4** XRD patterns of Sample B and montmorillonite powder.

differed appreciably in a wavenumber range of (1500–500 cm<sup>-1</sup>). In the IR spectrum of CI-WR, two new peaks occurring at 661 and 731 cm<sup>-1</sup>, were recorded, which was probably related to the stretching vibration mode of C–Cl bond in the chlorinated polymer. For example, the wavenumber at about 731 cm<sup>-1</sup> was assigned to C–Cl bond in the structure –CH<sub>2</sub>–CH<sub>2</sub>–CHCl– at the chain end.<sup>25</sup> The two new peaks proved that chlorine atom was grafted into WR molecular chain successfully.

Figure 3 presents the TG curves of WR and CI-WR within a temperature range from ambient to 800°C. The experimental data related to the two scans are provided in Table II, showing the temperatures for differing weight losses, i.e., T<sub>2%</sub>, T<sub>10%</sub>, T<sub>30%</sub>, T<sub>50%</sub>, and T<sub>70%</sub> and the char residue (%) obtained at 800°C. As shown in Figure 3, the decomposition of CI-WR in the temperature range of 170–320°C was fast and then slowed down considerably at the following temperature i.e., 320–800°C. The weight loss of CI-WR was about 62% at 800°C, suggesting that the decomposed component and char residue accounted for 62 and 38%, respectively. Moreover from Table II, it is also noticed that the char residue of CI-WR (38%) was much higher than that of WR (28%). This indicated that CI-WR had an excellent capability of promoting char formation at high temperature.

### Flammability behavior

The flammability behaviors of LDPE/CI-WR-Sb blends were evaluated with LOI,<sup>26,27</sup> and UL-94 tests.<sup>28,29</sup> Table III presents the resulting data for the LDPE/CI-WR-Sb blends at different mutual loading ratios. From Table III, we can see that the LOI values of LDPE were increased from 17.4 to 27.2 by the incorporation of CI-WR. This trend can be seen for Sample B where its LOI value (27.2) was much higher than that of self-extinguished polymer (26.0),<sup>30</sup> suggesting that CI-WR could be applied as a feasible flame retardant. Additionally, it is found that both Samples A and C have no rating in UL-94

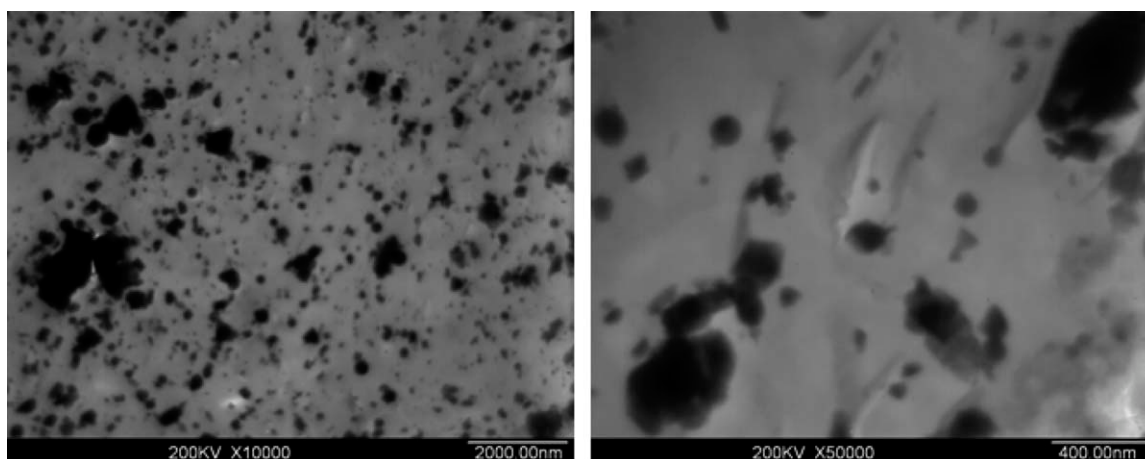


Figure 5 TEM images of Sample B at the magnifications of 10,000 and 50,000  $\times$ .

tests because of flaming dripping in the combustion, while Sample B reached to the highest grade, i.e., V-0 rating. By comparing the results of LOI values and UL-94 rating for Samples A, B, and C, it can be concluded that a proper mass ratio of CI-WR to  $\text{Sb}_2\text{O}_3$  is needed for a distinct synergistic effect.

#### Effect of flame retardant amount on the flame retardancy of LDPE/CI-WR-Sb

The amount of flame retardant had a significant influence on the flame retardancy and mechanical properties of LDPE/CI-WR-Sb blends. To optimize the content of the flame retardant in LDPE/CI-WR-Sb blends, the effect of different amount of flame retardant had been investigated. The LOI and UL-94 test data are shown in Table IV. It is noticed that the flame retardancy of the LDPE/CI-WR-Sb blends increased with the increasing flame retardant content. In regard to Samples D and E, the LOI values achieved were 24.9 and 25.6, respectively, with no

flame retardancy rating in UL-94. For Sample F, at 70 weight parts flame retardant per 100 LDPE weight parts, the LOI value reached up to 26.8 with a flame retardancy rating of V-1. More importantly, in case of Sample B, the combustion time ( $t_1$ ,  $t_2$ , and  $t_1 + t_2$ ) decreased extremely when the flame retardant content in the LDPE/CI-WR-Sb blends increased to 80 weight parts. This excellent flame retardancy implied that to achieve a V-0 rating, 80 phr is the minimum amount of the flame retardants that should be added to the LDPE matrix. For Samples G and H, although fire performances were satisfactory, polymer's mechanical properties would most probably be deteriorated concomitantly. Therefore, Sample B was thought to be the best choice for practical application in terms of balancing flame retardancy with mechanical properties of the LDPE/CI-WR-Sb blends.

#### Flame Retardant Mechanism

MMT plays an important role in improving the thermal stability and fire retardancy of the LDPE/CI-WR-Sb blends. This is because MMT has a good thermal stability and can migrate to the surface of the composites forming a glassy coating and stable carbonaceous charred layers at high temperature.<sup>31</sup> Therefore, to investigate the relationship between MMT and LDPE matrix is of significance. XRD and TEM are the two well-known techniques applied to characterize the micro-structure of MMT dispersed

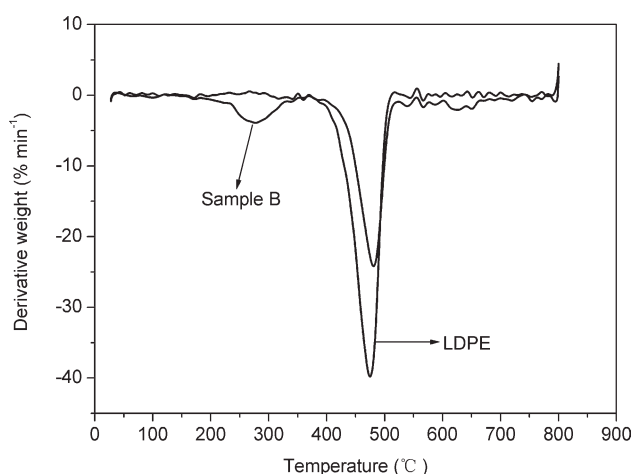
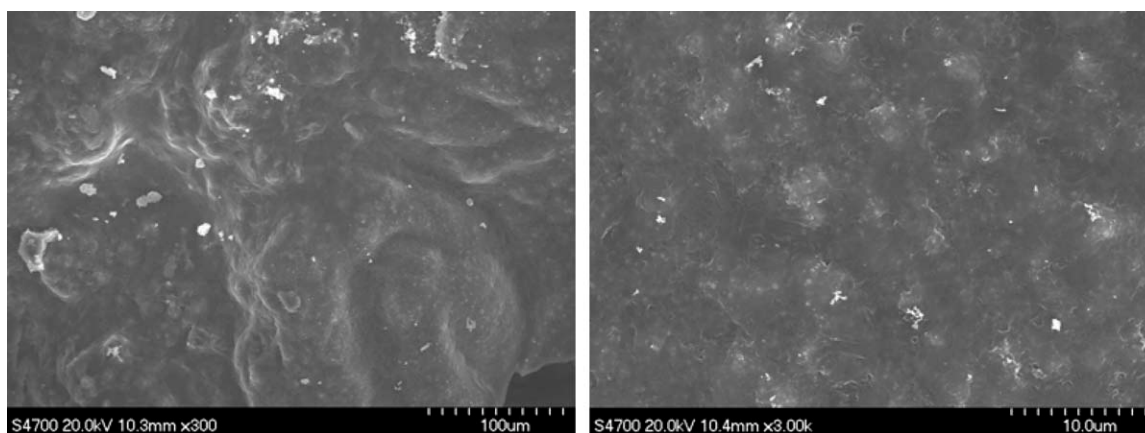


Figure 6 DTG curves of LDPE and LDPE/CI-WR-Sb blend of Sample B.

TABLE V  
Data of Thermal Degradation Temperature and Char Residue of Neat LDPE and LDPE/CI-WR-Sb Blends of Sample B

Sample No.	$T_{\text{initial}}$ (°C)	$T_{\text{max}}$ (°C)	Char residue at 800°C (%)
LDPE	442	475	3.5
B	241	482	12.8

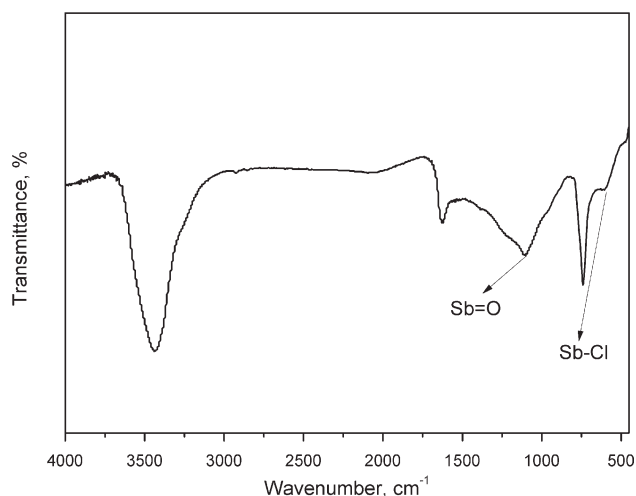


**Figure 7** Micrographs of the residue surfaces of Sample B after combustion at the magnifications of 300 and 3000  $\times$ .

in polymers. Figure 4 shows the XRD patterns of MMT powder and Sample B. There was an obvious peak shift for Sample B compared with the MMT powder, indicating that the interlayer spacing of MMT was significantly enlarged which implied the formation of intercalated structures in the composites of LDPE/MMT. The interlayer spacing of MMT could be calculated by the Bragg equation ( $2d_{001}\sin\theta = \lambda$ ,  $\lambda = 0.154$  nm). The MMT powder had a diffraction peak at  $2\theta = 3.17^\circ$ , corresponding to 2.8 nm of basal spacing, while Sample B had a peak at  $2\theta = 2.24^\circ$ , relating to 3.9 nm, with an approximate 40% enhancement. That was because LDPE chains were intercalated into the MMT galleries during the melting processing, and with the continued enlargement, there was no obvious peak in XRD patterns. Moreover, TEM provides another effective way to research the micro-structure of MMT in polymers optically. Figure 5 shows the TEM picture of Sample B containing MMT. The tiny white lines corresponded to MMT layers. It could be

seen clearly that MMT layers were fully oriented in the flow direction and were well-exfoliated in the LDPE composites (consistent with XRD analysis), forming a nanocomposite. Figures 4 and 5 demonstrate that an intercalated structure is formed in the composites of LDPE/MMT. The formation of the intercalated structures would improve thermal stability and flame retardancy of the LDPE/CI-WR-Sb blends.

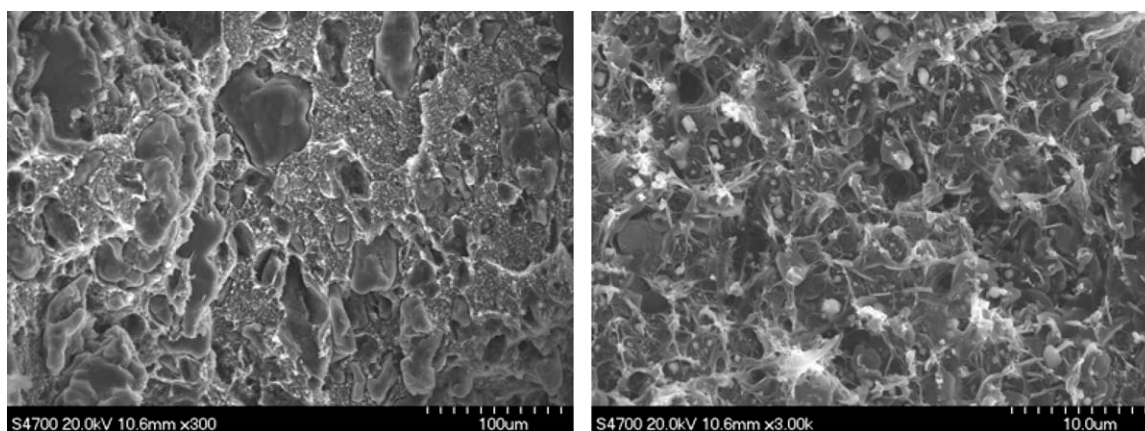
Differential thermogravimetric (DTG) curve is widely used to serve as useful indicators of polymer thermal stability and flammability.<sup>32</sup> Figure 6 indicates the DTG data of the neat LDPE and LDPE/CI-WR-Sb blend (Sample B) in nitrogen at a heating rate of  $10^\circ\text{C}/\text{min}$ . Figure 6 reveals that LDPE is thermally stable below  $400^\circ\text{C}$ , and at  $430^\circ\text{C}$ , it has a weight loss of less than 10% which almost exhausted at  $500^\circ\text{C}$ . Moreover, the decomposition rate of the neat LDPE was faster than that of Sample B in the temperature range of  $460$ – $800^\circ\text{C}$ . Table V lists the data from the DTG curves including the temperature of initial degradation ( $T_{\text{initial}}$ ), maximum rate of weight loss ( $T_{\text{max}}$ ) and the char residue at  $800^\circ\text{C}$ . It could be seen from Table V that  $T_{\text{initial}}$  of Sample B was lower than that of the neat LDPE. This was because CI-WR began decomposing at low temperature, generating some small molecules such as hydrochloric gas. The decomposition of CI-WR at low temperature provided a good protection to LDPE substrate. Moreover, it is also observed that the amount of char residue (12.8%) of Sample B was much higher than that of the neat LDPE (3.5%), which suggested that the former would promote the



**Figure 8** FTIR spectrum of the residual char after combustion for Sample B.

**TABLE VI**  
Data of Mechanical Properties of Neat LDPE and LDPE/CI-WR-Sb Blends

Sample No.	Tensile strength (MPa)	Elongation at break (%)
LDPE	9.3	477
B	8.4	110



**Figure 9** Micrographs of the cryo-fractured surfaces of Sample B at the magnifications of 300 and 3000  $\times$ .

char forming during combustion. This finding was consistent with the result depicted in Figure 3. It is believed that the char formation was responsible for the thermal stability and flammability properties.<sup>33,34</sup>

Figure 7 shows the SEM micrographs of the residue surfaces of Sample B after combustion. It is observed that the microstructures of the residue displayed a homogenous and compact structure, indicating the formation of a strong charred layer during combustion. The char formation may effectively prevent the heat and flammable volatiles transfer between the flame zone and the substrate, and thus resulted in good flame retardancy.

The FTIR spectrum of the residual char after combustion for Sample B is given in Figure 8. The broad peaks ranging from 1100 to 1250  $\text{cm}^{-1}$  and 540  $\text{cm}^{-1}$  were attributed to the stretching modes of  $\text{Sb}=\text{O}$  and  $\text{Sb}-\text{Cl}$  groups in the oxyhalide antimony ( $\text{SbOCl}_x$ ).<sup>35</sup> The FTIR spectra evidenced that an obvious synergistic effect existed during thermal degradation between CI-WR and  $\text{Sb}_2\text{O}_3$  in the LDPE/CI-WR-Sb blends. Thus we can conclude that the improved flame retardancy and thermal properties of the LDPE/CI-WR-Sb blends compared with the neat LDPE were attributed to char formation and synergistic effect between CI-WR and  $\text{Sb}_2\text{O}_3$  at high temperature.

#### Mechanical properties of LDPE and LDPE/CI-WR-Sb systems

Considering the fact that the LDPE/CI-WR-Sb blends of Sample B was the optimum recipe, thus mechanical properties of the neat LDPE and the blends were investigated and compared, as exhibited in Table VI. Mechanical properties were mainly involved in tensile strength and elongation at break. It is revealed that the tensile strength of Sample B (8.4 MPa) had a little decrease in comparison with that of the neat LDPE (9.3 MPa). What is worse, the elongation at break reduced sharply for Sample B

from 477 to 110% compared with the neat LDPE. This may be attributed to the lack of interfacial adhesion and poor compatibility between LDPE and CI-WR, as discussed elsewhere.<sup>36,37</sup> The above-mentioned observation is confirmed by the SEM microphotographs of Sample B, as shown in Figure 9. A wealth of visible voids existed in Sample B, which also indicated a bad compatibility between LDPE and CI-WR owing to their different polarity.

#### CONCLUSIONS

A new strategy of reclamation of WR was proposed by synthesizing a new flame retardant, CI-WR, via a water-based chlorination method. A suitable ratio of CI-WR to antimony trioxide could increase LOI value to 27.2 and pass UL-94 V-0 rating test for the LDPE/CI-WR-Sb blends. The good flame retardancy and the thermal properties of the LDPE/CI-WR-Sb blends are largely attributed to high char formation and distinct synergistic effect between CI-WR and  $\text{Sb}_2\text{O}_3$  at high temperature. The mechanical properties of the LDPE/CI-WR-Sb composites were slightly inferior to that of the neat LDPE due to the lower compatibility between LDPE and CI-WR.

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