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Influence of Novel Screw Elements on the Reactive Extrusion of Low Density Polyethylene grafted with Maleic Anhydride in Co-rotating Twin Screw Extruder

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Abstract: The influences of novel screw elements on grafting reactive extrusion for low-density polyethylene(LDPE) grafted with maleic anhydride(MAH) was investigated in Leistrize30/34 co-rotating twin screw extruder. The specimens were collected from three positions along the extrusion course and the die exit. The grafting degree (GD) of the specimens were determined by titration. In order to compare the influence of different screw elements on the grafting reactive extrusion, the experimental results of conventional screw element and kneading block were introduced. The results showed that different screw elements had remarkable influences on the grafting reactive extrusion and the grafting degree. The novel screw element including the S-type element with high elongation ability and the NI-MPE with back-flow action could improve the grafting degree rapidly. The kneading block with a good mixing performance had a higher grafting degree.

Keywords: Low density polyethylene grafted with maleic anhydride, Novel screw elements. Co-rotating Twin Screw Extruder.
PACS: 83.80Tc

INTRODUCTION

Polyethylene (PE) has been widely used in various fields for its excellent electrical properties, mechanical properties and processing performance. However, its application is restricted because of its poor heat resistance, printability, fire resistance and poor compatibility with other polar polymers and inorganic fillers. In order to improve the compatibility, grafting modification of PE is one of the most important means[1-4]. Maleic anhydride (MAH) melt grafting modified PE can significantly improve the PE bonding ability its compatibility with polar polymers [5-7]. Many researchers have done a lot of works, but there are few studies about the influence of reactive extrusion equipment, especially the new screw element on reactive grafting.

In this paper, the influence of novel screw elements on MAH grafted low density polyethylene (LDPE-g-MAH) in co-rotating twin-screw extruder was studied.

EXPERIMENTS

Materials

LDPE, LD100 - BW, produced by Sinopec Beijing Yanhua Petrochemical Co., Ltd. Chemical No. 1 Plant. MAH, analytical purity, produced by Beijing Yili Fine Chemicals Co., Ltd.. Dicumyl peroxide (DCP), chemical

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purity, recrystallization with anhydrous ethanol before use, produced by China Pharmaceutical Group Shanghai Chemical Reagent Company. Xylene, analytical purity, produced by Beijing Chemical Reagent Factory of Beijing Beihua Fine Chemicals Co., Ltd.. Acetone, analytical purity, produced by Beijing Century Red Star Chemical Co., Ltd..

Instruments and Equipment

LSM 30/34 intermeshing twin-screw extruder, manufactured by Leistritz, German. GH - 10 high speed mixer, manufactured by Beijing Plastic Machinery Factory. μPXRZ - 400A melt flow rate meter, manufactured by Science and Education Instrument Factory of Jilin University. DZX-1 vacuum drying oven, produced by Shanghai Fuma Laboratory Instrument Co., Ltd.. Nico let - 210 Fourier transform infrared spectrometer, manufactured by Nicolet, USA.

Preparation and Purification of Specimens

Weigh the material according to the proportion m(LDPE)/m(MAH)/m(DCP) is 100.0: 3.0: 0.1 and then pour them into a high speed mixer. After mixing for 2 minutes with the mixing speed is 2000 rpm, add the mixture to twin-screw metering feed hopper and melt-grafting in a twin screw according to the preset conditions. After the extrusion is stabilized, specimen online at a predetermined location (as shown in FIGURE 1). Promptly place the specimen in liquid nitrogen after sampling, to prevent further reaction during its cooling process. Each time a certain amount of melt grafted product is obtained, the screw elements (specific screw elements and their characteristics are shown in Table (1)) of the main reaction zone should be changed and then repeat the experiment. The process conditions of reactive extrusion are as follows: the barrel temperatures in the extrusion direction are 150, 160, 160, 160, 160℃; the die temperature is 150℃; the main screw speed is 30rpm; the feed speed is 60rpm, and the feed rate is 7.2kg/h.

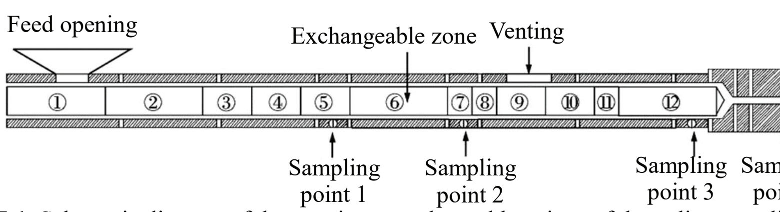


FIGURE 1. Schematic diagram of the reactive extruder and locations of the online sampling points

Note: ①② SE45/ 120 ③⑤ SE30/ 60 ④ SE20/ 60 ⑥ exchangeable zone (SE30/120 or TME or KB30/120 or NI - MPE) ⑦ SE30/ 30 ⑧ SE20/ 30 ⑨⑩ SE45/ 60 ⑪ SE45/ 30 ⑫ SE30/120

TABLE (1). Geometric parameters and characteristics of the novel screw elements

Name	Screw O.D./mm	Center Distance/ mm	Lead/ mm	Characteristic
SE30/120	34	30	30	Positive conveying element
TME	34	30		Distributing element
KB30/120	34	30		High shear element [8]
NI - MPE	34/30	30	30/60	Backflow element [9]
S-element	31	30	120	High elongation element

note: the staggered angle of KB30/120 is 30°

Add the extruded specimen to the three-necked flask filled with 150 mL of xylene, heating the oil bath to 140 °C and heated refluxing for 2 hours. After slightly cooling, filter the solution into unheated acetone. Prepare a pure grafted product specimen by precipitation and then suction filtration. The product specimen (filter cake) should be vacuum dried for 24 hours.

RESULTS AND DISCUSSION

The specimen after purification of the grafted product was compressed into a transparent film with its thickness of about 100 μm for FTIR measurement (as shown in FIGURE 2). As shown in FIGURE 2, the MAH characteristic absorption peak appears at 1790 cm-1, which indicates that MAH has been grafted onto the LDPE molecular chain.

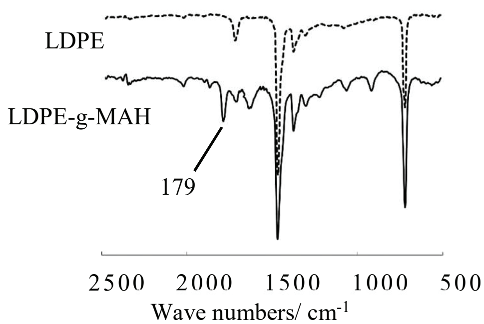


FIGURE 2. FTIR spectra of LDPE and LDPE - g - MAH

The G of specimens from each sampling point are shown in the TABLE (2).

TABLE (2). Grafting Ratios(%) of the Specimens

Exchangeable Zone Screw Elements	Sampling Position			
	Sampling Point 1	Sampling Point 2	Sampling Point 3	Sampling Point 4
SE30/120	7.045	14.007	25.768	53.040
KB30/120	8.976	18.989	34.227	68.220
TME	7.924	18.262	31.684	60.718
NI - MPE	0.633	13.023	25.260	50.520
S - element	5.398	17.873	27.903	58.127

As shown in Table (2), the G of specimen from sampling point 1 is KB30/120, TME, SE30/120, S, and NI-MPE in descending order. It is that the sampling point 1 is located in front of the screw exchangeable zone, thereof the G of specimen is directly affected by the pressure build-up capability of the screw elements in the subsequent exchangeable zone. Due to the non-intermeshing grooved screw element in the NI-MPE, clearance between the element and the inner wall of the barrel is large, so that the pressure is reduced. Therefore, the filling degree of the upstream flow channel is relatively low. The TME element is formed by the alternate combination of helical and straight teeth. The TME has no conveying capability, so the filling degree of the material in the upstream section is higher. The S – element combined both positive and negative conveying zone, has a certain positive conveying capacity, which decreases its filling degree. SE30/120 and KB30/120 have positive conveying capacities, which have strong pressure build-up capabilities.

In this work, the influence of the exchangeable zone element on G is illustrated by examining the relative increase in G at sampling point 2 and sampling point 1 (as shown in FIGURE 3 and TABLE (3)). The increase in specimen G is highest as the exchangeable zone is S - element, followed by the NI - MPE, TME third, KB30/120 and SE30/120 are lower. The main reason is that the mixing performance of each component and the filling degree of the segment are different. The S – element has stronger elongation performance due to its structure. The NI-MPE has better filling degree and backflow effect [10-11]. Many times of distribution and merging occurred when the material flows through the TME. However, due to geometric characteristics of KB30 /120, causes high shear flow of the material as it passes through the element, which greatly improves the dispersive mixing capability of the components and promotes grafting as well. Due to the stronger conveying capacity of SE30/120, the filling degree is lower relatively. Thus it can be seen that distributed mixing is more conducive to improving the G of product than dispersive mixing.

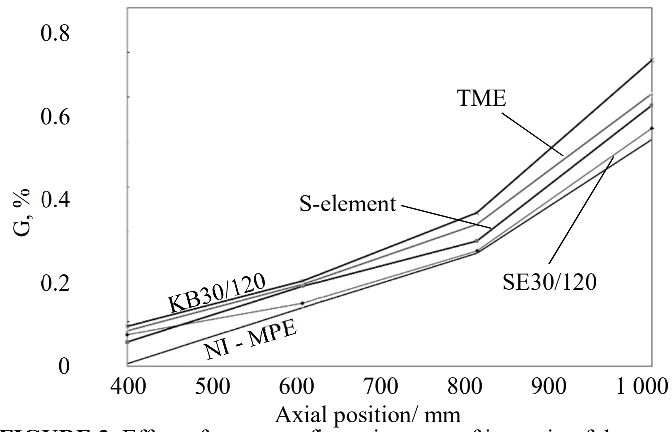
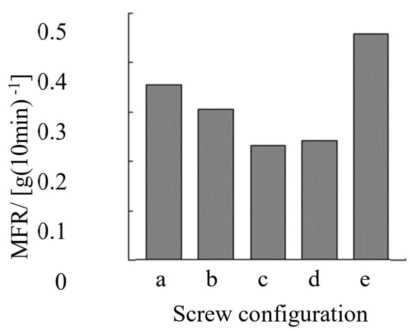


FIGURE 3. Effect of screw configuration on grafting ratio of the specimens

TABLE (3). Increase in G of the specimens taken from the sampling points before and after the exchangeable zone

G Increments of Exchangeable Zone Screw Elements	SE30/120	KB30/120	TME	NI - MPE	S - element
	0.069 623	0.100 125	0.103 385	0.123 898	0.124 369



a - SE30/120, b - S element, c - KB30/120；d - TME, e - NI - MPE

FIGURE 4. Effect of screw configuration on MFR of the end extrudate

In addition, as it also shown in FIGURE 3, all the G grows rapidly of configurations of specimens along the channel from the sampling point 3 to the sampling point 4, and the growth rate is about the same. The main reason is that the pressure of extruder die completely fills the melt between sampling point 3 and sampling point 4, which increases the grafting. This shows that the filling degree has a great influence on the G of the specimens.

The MFR of the purified grafted product was determined under the conditions of a load of 5 kg and a temperature of 190 °C.

The higher the G of final extruded graft product, the lower the MFR will be and vice versa. The conclusion shown in FIGURE 4 and TABLE (3) are in fair agreement.

CONCLUSION

As a high-shear element, the G of the specimen along the KB30/120 is the highest and the MFR is minimal. As a distributing element with a certain positive conveying capacity, TME makes the G of the specimen along the channel higher than that of the specimen G of the conventional screw element SE30/120. As a elongating element, S – element could improve the G of specimens.

In the melt-graft extrusion process, residence time, filling degree, and mixing performance of the screw elements have a greater impact on the specimen G. Distributive mixing is more conducive to improve the G of grafting product than dispersive mixing.

REFERENCES

1. DING Yonghong, CHENG Minlian, LIU Huaguo, et al. Functionalization Study on Melt-grafting Maleic Anhydride onto LLDPE [J]. Plastics Science and Technology, 2001, (6), pp. 1-5
2. Xu Xi, Cai Liaoyuan. Recent Advantages in Study of Modification and Fundamental Research Regarding of Polyolefi [J]. Modern Plastics Processing and Application, 1995, 7 (1), pp. 1-8
3. Bai Jingmei, Li Shucai. Grafting Maleic Anhydride onto LDPE with Melt Extrusion[J]. Plastics, 2005, 34 (2), pp. 53-55
4. Wang Yilong , Jian Xigao, Zhang Hongjin. Studying on Grafting Maleic Anhydride onto LDPE with Reaction Melt Extrusion [J]. China Plastics, 1993, 7 (1), pp. 34-38
5. Yin Jun, Zhang Jun. Characterization and Quantification of Maleic Anhydride Grafting Polyolefin[J]. Journal of Functional Polymers , 2002 , 15 (3), pp. 99-106
6. Chen Xiaoli, Li Binghai. Grafting of LLDPE with Maleic Anhydride by Reactive Extrusion[J]. Plastic , 2005 , 34 (6), pp. 6-9
7. Zhang Guangcheng. Study on Reactive Extrusion of Polyolefin [D]. Xi'an: Northwest Polytechnical University, 2001. 2
8. Zhang Junyi. Theoretical and Experimental Study of TME in Intermeshing Co-Rotating Twin-Screw Extrusion [D]. Beijing: Beijing University of Chemical Technology, 2003. 12
9. Machado A V , Covas J A , Duin M Van. A study of grafting reactions during processing of polyolefins [J]. Advances in Polymer Technology , 2004 , 23 (3), pp. 196-210
10. Chen Shihong. Theoretical and Experimental Study of NI-MPE in Intermeshing Co-Rotating Twin-Screw Extrusion [D]. Beijing: Beijing University of Chemical Technology, 2003. 3
11. Rosales C, Perera R, Ichazo M, et al. Grafting of Polyethylenes by Reactive Extrusion, I. Influence on the molecular structure [J]. Journal of Applied Polymer Science, 1998, 70 (1), pp. 161-176
12. Rosales C, Perera R, Ichazo M, et al. Grafting of Polyethylenes by Reactive Extrusion , II. Influence on rheological and thermal properties [J]. Journal of Applied Polymer Science , 1999, 73 (13), pp. 2 549-2 567