This technical paper was written and developed in October, 2001 when the author(s) was an employee of Dyneon LLC. Dyneon LLC was formerly a wholly-owned subsidiary of 3M Company and was fully integrated into 3M Company on January 1, 2011.

Title:
An Experimental Investigation of the Effect of Polymer Processing Additives (PPAs) on the Melting and Gel Formation Mechanisms in a Single Screw Extruder

Abstract:
Much work has been published showing how the addition of a polymer processing additive (PPA) has improved the processing characteristics of polyethylene. This paper will give insight on how the PPA affects gel formation in linear low density polyethylene (LLDPE) and polyolefin plastomers (POP) in a single screw extruder. Solids conveying, melting, and melt conveying were experimentally studied to determine how the

Date Published: October, 2001
An Experimental Investigation of the Effect of Polymer Processing Additives (PPAs) on the Melting and Gel Formation Mechanisms in a Single Screw Extruder

K. R. Slusarz
Davis-Standard Corporation

J. P. Christiano
Davis-Standard Corporation

Stephen E. Amos
Dyneon LLC


ABSTRACT

Much work has been published showing how the addition of a polymer processing additive (PPA) has improved the processing characteristics of polyethylene. This paper will give insight on how the PPA affects gel formation in linear low density polyethylene (LLDPE) and polyolefin plastomers (POP) in a single screw extruder. Solids conveying, melting, and melt conveying were experimentally studied to determine how the PPA affected these important processes during extrusion leading to improvements in the melt quality.

INTRODUCTION

Changes in molecular architecture of polyethylenes have introduced new materials that have unique physical properties. Along with enhanced properties of the end products, these materials have very different processing characteristics. It has been shown that modification to single screw design or processing conditions may be necessary in order to process these new families of resins1,2,3.

The primary use of PPA has been the elimination of melt fracture in polyethylene films1. Other processing additive benefits such as widening processing windows, improvement in surface properties4, and eliminating die lip build-up5 have also been demonstrated. A new benefit of gel reduction has also been identified and several mechanisms postulated6. This work will show that certain PPA products can reduce unmelted gels by preventing the premature, frictional melting of plastomers in the feed section of the extruder.

A PPA forms a thermally stable fluoropolymer coating on the metal surfaces of the extruder. A properly designed PPA will move to the area of highest shear. This is typically the metal die surfaces and where the screw flights sweep tightly across the extruder barrel. The fluoropolymer coating changes the interfacial properties between the metal surface and the host resin – preventing stick and lowering frictional forces. It is through this interfacial mechanism that a plastomer can be extruded without premature melting in the feed section.

This paper will look at the changes that occur with the addition of PPA on solids conveying, melting characteristics and process performance. It will investigate how PPAs may allow a more general purpose screw design to run a wider spectrum of materials with good quality.

EXPERIMENTAL

Experimentation was conducted on two materials, a 3 M polyolefin plastomer (POP) and a 3 M Zigar-Natta catalyzed linear low density polyethylene (LLDPE). In addition three different PPAs were evaluated, PPA 1 (Dynamar™ FX 9613), PPA 2 (Dynamar™ FX 5920A) and PPA 3 (Dynamar™ FX 5911X). The POP has been shown to have early melting behavior7. This paper will focus on the solids conveying and melt pool formation of these materials and how they are affected by PPAs.

Extrusion tests were conducted on a 63.5 mm 24:1 L/D extruder with pressure transducers installed along the length of the barrel. The transducers were placed at 3 L/D intervals starting at 3.8 L/D from the feed opening. The pressure was measured at a sampling rate of 50 Hz per transducer. The melt temperature was recorded at the discharge of the extruder using an exposed junction melt thermocouple immersed to the center of the melt stream. All of the tests were conducted using a 41 cm wide film die with a die gap of 1.1 mm.

All of the tests were conducted on a barrier screw that was optimized for cast film running LLDPE. The barrier screw had a secondary flight introduced in the transition section of the screw. This secondary flight had a clearance greater than the main flight (barrier gap). This flight separated the solid bed from the melt pool, while the barrier clearance allowed the melted polymer to pass from the solids channel to the melt channel. The lead of the main flight within the barrier section was increased in order to maximize the available area for melting against the barrel wall. The barrier flight had a lead greater than the main flight, which changed part way through the barrier section. This decreased the volume of the solids channel while increasing the volume of the melt channel as the polymer proceeded down the barrier section of the screw. In addition to changing the width of each channel, the channel depths were altered. The depth of the melt channel was increased, while the depth of the solids channel was decreased, as they progressed down the screw, in order to match the volume of the melted polymer. The barrier gaps and channel depths that were selected are typical of those used in production cast film screws. The screw had an overall compression ratio of 2.53:1 and incorporated a Union Carbide mixing section in the meter section of the screw to ensure melt homogeneity.

The test matrix included two cast film resins, a control (LLDPE) and one known to have early melting characteristics (POP), and three different PPAs. The barrel profile was kept constant for all tests at: Zone #1 - 177°C (350.6°F), #2 - 204°C (399.2°F), #3 - 232°C (449.6°F), #4 - 260°C (500°F), while the die and adapter temperatures were kept at 260°C (500°F). The breaker plate pressure was fixed at approximately 20 MPa by a valve in the adapter. All of these conditions are typical for running production LLDPE cast film. Overall performance was characterized by specific output, melt temperature, power consumption, total melt temperature variation, and total pressure variation. The solids conveying, melting, and pumping processes within the extruder were monitored and interpreted by means of pressure traces which were recorded at very high sampling rates8. The material was extruded into a roll and examined visually for melt quality (gels – size and quantity).

The PPAs were added to the materials by means of a...
the pressure range at each pressure transducer. Figure #6 represents the nominal pressure profile, while the box shows gradient. Figure #5 shows the pressure development along pressure profile along with a lower cross channel pressure Figure #4. The addition of PPA caused a reduction of the reducing the amount and size of gels in the extrudate, see stability of the system. It did improve the extrudate quality by the addition of PPA 2 to the POP did not change the process screw speed is shown in Figure #3. This will be discussed in greater detail in the Pressure Analysis section. The process stability was considered excellent for all the materials extruded with and without PPA 2 at a screw speed of 50 RPM. The resins were blended with 0.15% blue masterbatch, which was in a LLDPE carrier, in order to observe the flow fields and melt pool generation.

**RESULTS AND DISCUSSION**

The results and discussions will focus on the interaction of PPA 2 and the two materials since this additive was specifically designed for film grade POP and LLDPE resin containing antiblock. Typically these applications have the most sensitivity to gel formation. The other two PPAs were also effective in POP and LLDPE and were tested to compare their effectiveness for unmelt gel formation.

**Process Performance**

The addition of PPA 2 to both materials showed a reduction in the output rate (Figure #1) and power consumption (Figure #2), by approximately 5%. The melt temperature versus screw speed is shown in Figure #8. This will be discussed in greater detail in the Pressure Analysis section. The process stability was considered excellent for all the materials extruded with and without PPA. The total melt temperature variation was less than 0.2°C (36°F), while the total pressure variation measured in the adapter pipe remained below 0.7%.

**Pressure Analysis**

The addition of PPA 2 to the POP did not change the process stability of the system. It did improve the extrudate quality by reducing the amount and size of gels in the extrudate, see Figure #4. The addition of PPA caused a reduction of the pressure profile along with a lower cross channel pressure gradient. Figure #6 shows the pressure development along the screw for the POP material at 50 screw RPM. The line represents the nominal pressure profile, while the box shows the pressure range at each pressure transducer.

The analyses of the screw carcasses or melt pool formation are presented in Figure #7. This graph shows the width of the solid bed (X) over the total width of the bed (W). As the figure demonstrates, the POP without PPA initiates melting approximately 4 L/D down the screw, which is still in the feed section of the screw. The addition of PPA delays the start of melting until just after 5 L/D down the screw, at the start of the barrier section. The amount of melt accumulation was always less for the POP with PPA as compared to the virgin POP. This helps explain the lower output rates and power consumption seen with the addition of PPA. The PPA had a similar effect on the LLDPE where the onset of melting and the melt pool accumulation are less for the PPA containing material. These results are consistent with other work in which the PPA was incorporated into the resin directly from the resin supplier. The pressure profile, onset of melting, and melt pool formation were all reduced in the resin containing PPA.

The PPA helped eliminate the unmelted gels by delaying the onset of melting until the introduction of the barrier section. Additionally, the PPA reduced the size of the melt pool in the solids channel, thereby preventing the solid bed from breaking up. This breakup causes unmelted material to be encapsulated in the melt creating gels in the extrudate. Figure #8 shows a pressure trace before and after PPA #2 was added to the POP at 50 RPM. The variation, which was attributed to solid bed breakup, was reduced after the PPA coated the extrusion system. It has been postulated that PPA coating helps eliminate gels caused by thermal and/or oxidative degradation. In coating the screw and barrel it prevents material from stagnating and creating gels at these surfaces.

**Additional PPA**

The other PPAs performed similarly to PPA #2 with regards to pressure, temperature, output and reducing unmelt gels.

The POP material, which has been shown to have a very high melting rate along with early melt pool formation, ran with acceptable process stability but had a significant amount of gels in the extrudate. Prior work showed that these gels were formed from unmelted material being entrapped in the melt due to solid bed breakup. By altering the starting point of the barrier section they were able to minimize the amount of solid bed breakup and match the screw design to the melting rate of the material. The addition of PPA was able to achieve the same results by delaying the start of melting until the barrier section was developed.

**CONCLUSIONS**

The addition of PPA can delay the onset of melting, lower the pressure profile, and improve melt quality (less gels) of POP when processed on an LLDPE barrier type screw. The PPA can cause output rates to drop slightly at the same RPM but.
higher specific outputs are possible with PPA resin run at the same head pressure. The PPA gives the processor the ability to alter the melting characteristics of POP in order to utilize a more general purpose screw design. Screw design changes can yield similar benefits1, reducing or eliminating the need for PPA addition.

REFERENCES


ACKNOWLEDGEMENTS

We would like to thank Tom Butler of the Dow Chemical Company and Susan Woods of Dyneon LLC for their contributions of materials and expertise.
**Figure #3** – Melt Temperature vs. Screw Speed for LLDPE and POP with and without PPA 2

**Figure #4** – Gel Response of POP with Addition of PPA

**Figure #5** – Pressure Profile - POP at 50 RPM
Keywords: LLDPE, Barrier Screw Design, Film, Polymer Processing Additive
Technical Information and Test Data

Technical information, test data, and advice provided by Dyneon personnel are based on information and tests we believe are reliable and are intended for persons with knowledge and technical skills sufficient to analyze test types and conditions, and to handle and use raw polymers and related compounding ingredients. No license under any Dyneon or third party intellectual rights is granted or implied by virtue of this information.

Important Notice:
Because conditions of product use are outside Dyneon’s control and vary widely, user must evaluate and determine whether a Dyneon product will be suitable for user’s intended application before using it. The following is made in lieu of all express and implied warranties (including warranties of merchantability and fitness for a particular purpose): If a Dyneon product is proved to be defective, Dyneon’s only obligation, and user’s only remedy, will be, at Dyneon’s option, to replace the quantity of product shown to be defective when user received it or to refund user’s purchase price. In no event will Dyneon be liable for any direct, indirect, special, incidental, or consequential loss or damage, regardless of legal theory, such as breach of warranty or contract, negligence, or strict liability.

© Dyneon 2001
Issued: 10/01
Printed in USA
98-0504-1402-2
All Rights Reserved

Product Information:
+1 651 733 5353   +1 800 723 9127

Dyneon Sales Offices:
Dyneon LLC
6744 33rd Street North
Oakdale, MN 55128
Phone: +1 651 733 5353
Fax: +1 651 737 7686
Toll Free: +1 800 723 9127

Dyneon GmbH & Co. KG
Hammfelddamm 31
D-41460 Neuss, Germany
Phone: +49 2131 14 2227
Fax: +49 2131 14 3857

Dyneon GmbH & Co. KG
Werk Kelsterbach
D-65444 Kelsterbach, Germany
Phone: +49 6107 772 516
Fax: +49 6107 772 517

Dyneon N.V.
Canadastart 11, Haven 1005
B-2070 Zaventem, Belgium
Phone: +32 3 250 7537
Fax: +32 3 250 7905

Dyneon GmbH & Co. KG
Succursale France
Boulevard de l’Oise, Tour 3M F-95500 Cergy
France
Phone: +33 1 3031 6611
Fax: +33 1 3031 6613

Dyneon GmbH & Co. KG
Sede Secondaria Italia
Via San Biolo, 3
Milano San Felice
I-20090 Segrate (MI) Italy
Phone: +39 02 7035 3206-7
Fax: +39 2 7035 3208

www.dyneon.com

Dynamar and Dyneon are trademarks of Dyneon
Warranty, Limited Remedy, and Disclaimer: Many factors beyond 3M’s control and uniquely within user’s knowledge and control can affect the use and performance of a 3M product in a particular application. User is solely responsible for evaluating the 3M product and determining whether it is fit for a particular purpose and suitable for user’s method of application. Unless a different warranty is specifically stated in the applicable product literature or packaging insert, 3M warrants that each 3M product meets the applicable 3M product specification at the time 3M ships the product. 3M MAKES NO OTHER WARRANTIES OR CONDITIONS, EXPRESS OR IMPLIED, INCLUDING, BUT NOT LIMITED TO, ANY IMPLIED WARRANTY OR CONDITION OF MERCHANTABILITY OR FITNESS FOR A PARTICULAR PURPOSE OR ANY IMPLIED WARRANTY OR CONDITION ARISING OUT OF A COURSE OF DEALING, CUSTOM OR USAGE OF TRADE. If the 3M product does not conform to this warranty, then the sole and exclusive remedy is, at 3M’s option, replacement of the 3M product or refund of the purchase price.

Limitation of Liability: Except where prohibited by law, 3M will not be liable for any loss or damages arising from the 3M product, whether direct, indirect, special, incidental or consequential, regardless of the legal theory asserted, including warranty, contract, negligence or strict liability.

Technical Information: Technical information, recommendations, and other statements contained in this document or provided by 3M personnel are based on tests or experience that 3M believes are reliable, but the accuracy or completeness of such information is not guaranteed. Such information is intended for persons with knowledge and technical skills sufficient to assess and apply their own informed judgment to the information. No license under any 3M or third party intellectual property rights is granted or implied with this information.