

Mineral reinforcement of linear-low density polyethylene film, bags, and liners

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ABSTRACT: *Specific modifications to the blown film processing and physical properties of butene, hexene, and octene copolymer linear low-density polyethylene (LLDPE) bags and liners are examined with the use of zero to 20% calcium carbonate mineral reinforcement. Increased film ductile strength is obtained with all three resin types without the loss of yield/stiffness properties critical to thin film conversion characteristics and end-use performance. Improved thermal and rheological properties of the resulting polyethylene composites yield processing benefits including increased extrusion efficiency, output rate, and bubble stability. These improvements have allowed the replacement of conventional low-density polyethylene (LDPE) as a blend component for improving LLDPE extrudability and bubble stability, without the MD/TD property anisotropy of blown film LDPE/LLDPE blends.*

KEYWORDS: *Calcium carbonate, copolymers, film, linear low density polyethylene, liners, minerals, physical properties, plastic bags.*

This paper discusses the results of a series of blown film extrusion and bag conversion trials conducted at the Heritage Bag Co. facility in College Park, GA, utilizing concentrates manufactured at the Heritage Plastics, Inc. compounding plant in Pica-yune, MS.

A previous paper (1) detailed the fundamental changes in the thermal and rheological properties of linear-low-density polyethylene (LLDPE) with calcium carbonate incorporation. The addition of mineral to the polymer increases thermal conductivity, and re-

duces the heats of fusion and crystallization of the composite. These two factors improve the thermal efficiency of the film extrusion process by increasing the rate of heat transfer and reducing the amount of heat which must be transferred into and out of the extrudate during melting and subsequent bubble cooling.

Rheologically, the addition of calcium carbonate to LLDPE increases its extensional viscosity and therefore its bubble stability during the blown film process without the development of strain hardening.

Results and discussion

Resins and minerals evaluated

The physical properties obtained with calcium carbonate mineral reinforcement of LLDPE films are dependent on several mineral and polymeric factors. Mineral factors include particle size distribution, which encompasses the average particle size (APS), top cut or maximum particle size, and the packing fraction. For the reinforcement of polyethylene, the surface must be treated to be rendered lipophilic and hydrophobic, so that the mineral may be "wetted" by the polymer.

LLDPE resin factors will play a major role in the properties obtained with mineral reinforcement. These factors include comonomer type (1-butene/1-hexene/1-octene), polymer molecular weight and molecular weight distribution (MW/MWD), polymer density/crystallinity, and polymer short-chain branching distribution.

For this study, commercially available LLDPE resins of each comonomer type (Et/Bu, Et/Hx, Et/Oc) were selected. All three are relatively narrow MWD film-extrusion grade materials of 1.0 MI. Being in the 918–920 kg/m³ density range, they are similar in crystallinity. While the 1-butene and 1-hexene copolymers are gas-phase reactor products, the 1-octene copolymer is a solution reactor product, and may therefore differ slightly in total crystallinity and short-chain branching distribution.

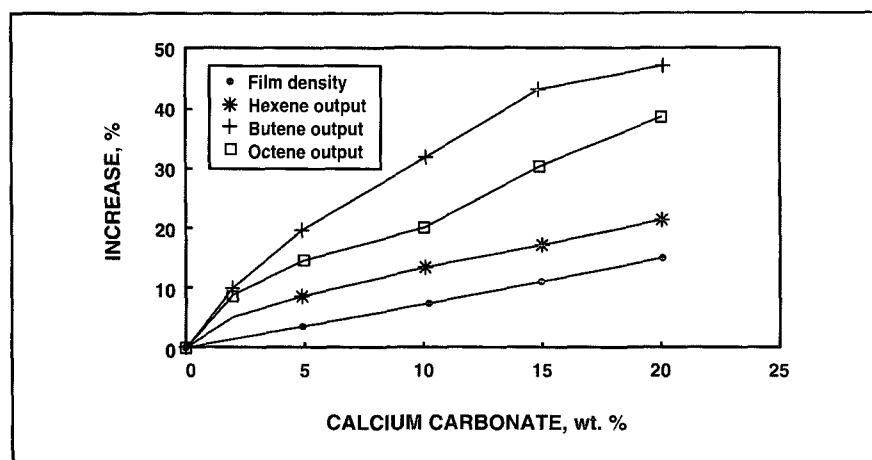
The effects of increasing mineral reinforcement level were determined in the range of 0% to 20% by weight (0–7% by volume) for each LLDPE resin. Fine-ground calcium carbonate (1 mi-

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I. Effect of mineral reinforcement concentrate addition to LLDPE resin on blown film processing conditions. 70 mm 24/1 L/D extruder at 115 rpm, 230 mm die with 1.4 mm die gap: film at 15 μ m, 3.4:1 BUR converted into 610 mm x 584 mm can liners

% Mineral	0	2.5	5	10	15	20
Butene copolymer						
Motor current A	63	67	69	70	72	72
Melt pressure, MPa	34.4	35.1	35.8	35.8	35.8	35.8
Melt temperature, °C	216	216	215	214	214	214
Output rate, kg/h	50.0	54.9	60.2	65.9	71.8	73.6
Hexene copolymer						
Motor current, A	73	73	72	71	71	70
Melt pressure, MPa	36.2	35.8	35.7	35.4	35.4	35.4
Melt temperature, °C	226	225	224	223	223	222
Output rate, kg/h	68.3	71.7	74.1	77.4	80.0	83.0
Octene copolymer						
Motor current, A	84	82	82	81	78	77
Melt pressure, psi	19.3	19.3	19.0	18.6	17.9	16.5
Melt temperature, °C	219	219	219	218	217	216
Output rate, kg/h	74.9	81.4	85.9	90.0	97.7	104

1. Density and output rate increase comparison



cron average particle size, 8 micron top-cut), treated with stearic acid to change the particle surface from lipophobic to lipophilic, was incorporated into all films via a 75% by wt. concentrate utilizing a higher alpha-olefin LLDPE concentrate carrier.

Effects of mineral reinforcement on the film extrusion and conversion process

Films of 15 μ m (0.6 mil) and 3.4:1 blow-up

ratio were extruded and in-line converted into 610 mm x 584 mm industrial can liners on a 70 mm, 24/1 L/D extruder fitted with a 230-mm die and 1.4-mm gap. Table I summarizes the effect on extrusion conditions with the addition of 0–20% calcium carbonate to the three LLDPE resin types.

Each comonomer type LLDPE displayed increases in output rate with the addition of calcium carbonate. These increases were beyond that expected due to the increase in melt density

imparted by mineral incorporation, as shown in Fig. 1. Addition of 20% calcium carbonate to LLDPE increases the composite density from 0.92 to 1.060, or 15%. Yet extrusion output increased by +22% for the hexene copolymer (68.3 $\pm$ 83.0 kg/h), +39% for the octene copolymer (74.9 $\pm$ 104 kg/h), and +47% for the butene copolymer (50.0 $\pm$ 73.6 kg/h). The film sealing conditions were not altered during these extrusion and conversion trials.

Film property modifications with mineral reinforcement

Film optical properties

As expected, the addition of calcium carbonate to LLDPE increases film haze and opacity, and reduces film gloss. While this limits the use of CaCO₃ in clarity applications, it will serve as an advantage in pigmented films by increasing opacity.

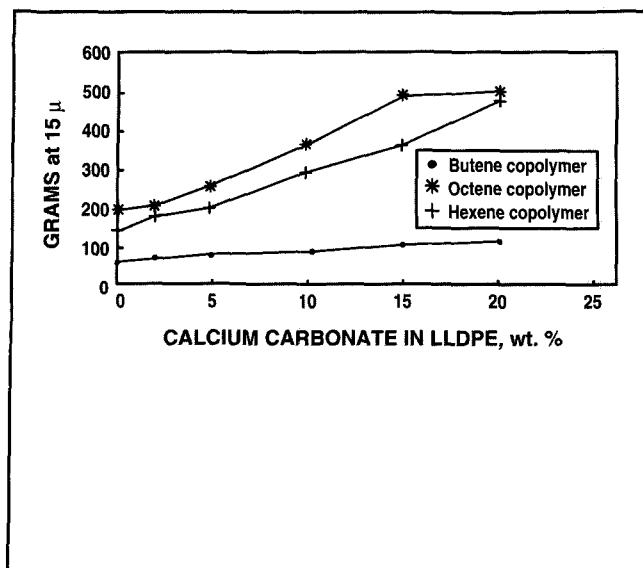
Film mechanical properties

Figure 2 displays the effect of 0–20% addition of calcium carbonate on the falling dart impact strength (ASTM D 1709) of the three types of LLDPE films. Improvements in dart impact strength are observed, regardless of comonomer type. While the impact strength of the butene copolymer LLDPE increases from 59 g to 104 g with 20% CaCO₃ addition, the higher alpha-olefin resins (1-hexene and 1-octene) display most dramatic improvements in ductile strength.

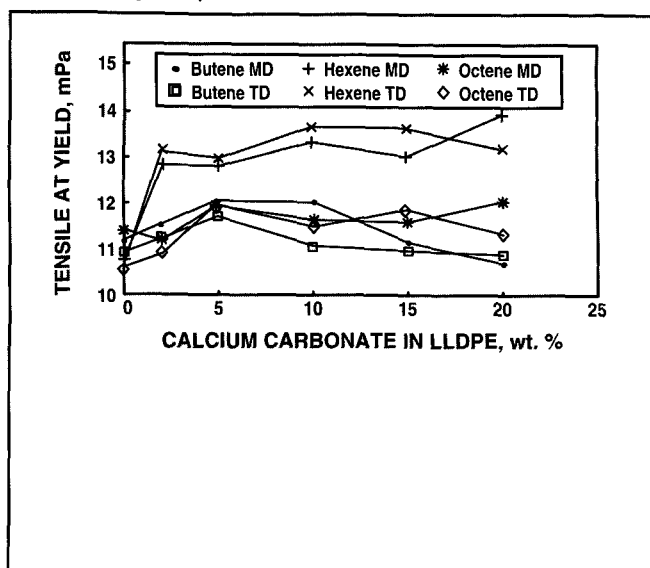
Research conducted by Fu and Wang (2) on the reinforcement of high-density polyethylene with treated calcium carbonate discusses the mechanisms of rigid inorganic particle toughening, and helps explain these dart impact improvements and the reasons for the difference in impact improvement between butene and higher alpha-olefin LLDPE copolymers.

Fu and Wang propose that cavitation and crazing of the polymer matrix surrounding the rigid, surface-treated

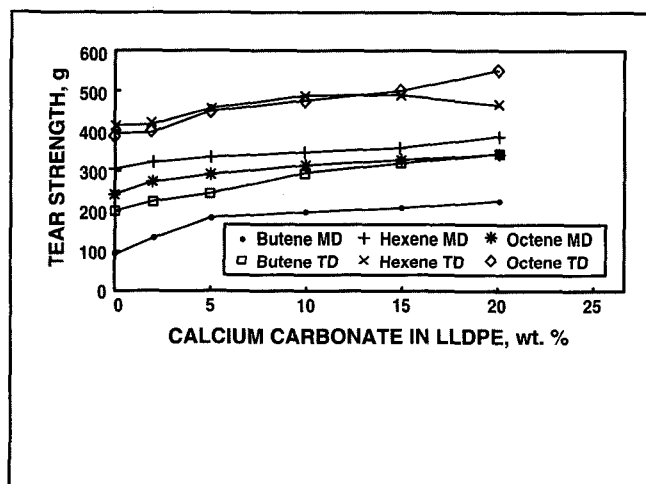
2. Dart impact response to mineral reinforcement



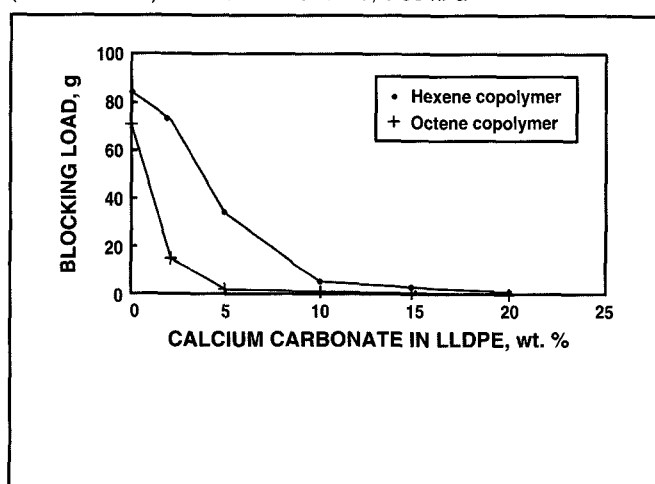
3. Yield strength response to mineral reinforcement



4. Tear strength response to mineral reinforcement



5. Film blocking response to mineral reinforcement. Film blocking load (ASTM D 3354). Induced 24 h at 60°C, 0.96 kPa



calcium carbonate particles allows the release of deformational stresses, allowing the composite to yield with the absorption of a great amount of energy. The dissipation of this energy over many finely dispersed particles is what toughens the polymer/mineral composite.

Once cavitation has occurred because of polymer-particle separation, the tear strength of the polymer matrix will determine the amount of energy absorbed during further strain. The higher alpha-olefin copolymers, with their much higher tear strength, will offer much greater resistance to the

propagation of these cavities once formed in the composite. This allows more energy to be absorbed from the falling dart prior to film rupture.

The tensile strength at film yield (ASTM D 882), which is directly related to the load capacity of a bag or liner at a given film thickness, is not significantly changed over this range of mineral loading (Fig. 3). This is critical as losses in film yield would require increases in film thickness in order to maintain the load-carrying capacity of a given liner.

The response of film propagation tear resistance to mineral concentrate

addition, as measured by the Elmendorf tear resistance (ASTM D 1922), is shown in Fig. 4.

Mineral reinforcement effects on bag and liner end-use performance

Openability of a bag or liner is a critical aspect of its performance, both in terms of avoiding product damage during end-use, and the negative impact on quality a blocked bag has on the customer. This performance may be quantified utilizing test methods such as ASTM D 3354, Blocking Load of Plastic Film by the Parallel Plate Method (3).

Figure 5 displays the blocking load of the 1-hexene and 1-octene copolymer films after blocking was induced at 60°C for 24 h at 0.96 kPa pressure. At the 15% by wt. mineral level the instrument utilized was at the limit of its resolution with both resin types. At 20% by wt. mineral, no film blocking could be induced with either resin type.

Conclusions

The application of calcium carbonate mineral reinforcement has been observed to improve the efficiency of blown film extrusion, yielding increases in process output in excess of that expected due to increased melt density alone. This increased performance is most likely due to increased melting and pumping efficiencies of the composites resulting from the thermal and rheological modifications discussed previously.

Film opacity is increased with mineral addition, which may allow the reduction of pigment level. The increased melt extensional viscosity imparted by mineral incorporation may allow the replacement of conventional LDPE in resin blends as a bubble stabilizing component where optical properties are not critical.

Thin films may be produced at high output rates without the development of MD/TD property anisotropy, as is experienced with the addition of conventional branched low density polyethylene to LLDPE for bubble stabilization (4).

Proper application of mineral reinforcement has demonstrated the ability to improve film ductile strength without concurrent loss in film tensile yield strength. This is critical to bag and liner performance where improved ductility increases film resistance to local failure, but where a loss in tensile yield strength would reduce the bag load capacity at a given thickness.

The higher alpha-olefin based films show no significant change in Elmendorf tear strength over the mineral loading range studied. The tear strength of the 1-butene copolymer film increased with concentrate addi-

tion, most likely due to the use of an HAO-LLDPE resin as the concentrate carrier.

While not necessary during these tests, the bag sealing process may have to be altered with increased mineral loading to compensate for changes in thermal properties. Depending on film type and thickness, bagmaking equipment, and operating conditions, the improved thermal conductivity of the film may require decreases in sealing temperature, pressure, and/or dwell time.

Bag and liner end-use performance are enhanced by improvements in film blocking resistance and bag openability with a minimum of 5% by wt. calcium carbonate reinforcement. □

Literature cited

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