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DEPARTMENT

Olefin Plastics

TITLE

Survey of Fillers and Reinforcements for Polyethylene

9

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IN FULL
REPORT

PLAINTIFF'S EXHIBIT

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This
report
is:☐ INTERIM
☒ FINALand mainly: ☒ NEW
☐ REVIEWDESCRIPTIVE SUMMARY
WITH CONCLUSIONS:

(Include in this space references to data books, and to earlier related reports, patents and publications.)

Summary:

Several fillers and reinforcements for polyethylene were discussed along with the properties of the filled resins. The fillers discussed include silica fillers, calcium carbonate, talc, fiberglass, asbestos fibers, ZnO, wood flour calcium sulfite, and calcium sulfate.

Conclusions:

1. Due to the large varieties of fillers, and the variability of a particular filler (particle size, surface treatment, etc.) a filler system needs to be evaluated with each resin to determine processability and end use properties.
2. In general fillers impart the following properties to polyethylene:
 - a. Greater stiffness
 - b. Reduced mold shrinkage
 - c. Lower melt index and higher melt viscosity
 - d. Decreased tensile and impact strength
 - e. Increased density
 - f. Improved creep and heat distortion temperature

Discussion:

Fillers may be divided into three groups: low cost fillers (extenders), functional fillers, and reinforcements.¹ An extender decreases the cost of a plastic material while a functional filler contributes a particular property to a plastic material such as improved electrical insulation. A reinforcement such as glass fibers, asbestos fibers, carbon fibers, etc. are added to improve the strength and may increase the cost of a plastic material. In general a filler imparts the following properties to a plastic material:

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- a. Greater stiffness
- b. Reduced mold shrinkage
- c. Lower effective melt indexes
- d. Higher melt viscosity
- e. Decreased tensile strength
- f. Increased density
- g. Lowered impact resistance
- h. Improved creep
- i. Improved heat distortion temperature

Several commercial types of fillers have been used in polyethylene^{2,3,4,5,6,7,9,10,13}

- a. Silica fillers, Kaolin clays
- b. Calcium carbonates
- c. Talcs
- d. Fibers such as glass, asbestos, etc.
- e. Metal oxides, MgO, ZnO
- f. Wood flour
- g. Calcium sulfate and calcium sulfite.

Silica fillers such as Superfloss increase the stiffness of polyethylene with the greatest increase in stiffness corresponding to the smallest particle size of the filler. The silica fillers also reduce the impact strength, tear resistance, reduces mold shrinkage and degrade the electrical properties.^{2,5} Calcium carbonate increases the stiffness, lowers the tensile strength, lowers the impact resistance, lowers the tear resistance of films, reduces mold shrinkage and improves opacity.^{2,4,7} Talcs increase the stiffness, decrease impact strength by a smaller amount than silicas and calcium carbonate, increases melt tension, decreases tear resistance of film and reduces mold shrinkage.^{2,4,7}

The fiber materials, asbestos and glass give good mechanical properties to polyethylene. Asbestos imparts increased stiffness, lower tensile strength, and lower tear strength of films. Asbestos fibers, however, have been associated with lung cancer and the asbestos requires special handling equipment. Glass fibers reduce mold shrinkage, increase stiffness, improve heat resistance, and increase tensile strength.^{4,6} The impact strength of polyethylene filled with glass seems to depend on the resin used. Some resins show improved impact strength with glass while the impact strength of some resins remains unchanged, and still other resin exhibit reduced impact strength when filled with glass fibers.^{6,11} Fiber-glass should be "sized" with a suitable coupling agent for good wetting and bonding of the polyethylene to the glass fiber. A typical "sizing" agent is an organic silane. Zinc oxide compounded into polyethylene has been reported to impart increased hardness and stiffness without affecting melt flow and toughness.⁸ Another investigator has shown that zinc oxide lowers the melt index, increases stiffness and increases melt tension.⁹ Wood flour in

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concentration of 0 to 50% has been used to fill polyolefins. The wood flour is reported to improve impact resistance and humidity tolerance of polyolefins.¹² An extruder with a vacuum part for moisture removal is necessary for compounding with wood flour. Wood flour normally has a high enough moisture content to produce severe foaming in the resin. Lion Fat and Oil Company (Japan) have patented the use of calcium sulfite and calcium sulfate as fillers for polyolefins. They report improved flexural and tensile strengths and non-flammability with the use of these fillers.¹³

In terms of cost per pound, the fillers listed above rank in cost from highest to lowest as follows:^{9,10} Zinc oxide>fiberglass>asbestos>silica>Kaolin clay>talcs>calcium carbonate. It is important to achieve good wetting of the filler by the polymer when compounding. The wetting is related to melt viscosity, processing pressure and processing temperature.¹¹ In general, the processing pressure and temperature of a filled polymer is higher than that of an unfilled polymer. To complicate matters, a particular filler may be obtained in different forms, particle sizes, surface treatment, etc., and these differences will affect the properties and processability of the filled polymer. Therefore, any filler system needs to be evaluated on the particular resin in question to determine the proper means of processing and to check desired end use properties.

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Table I

Effects of Fillers on Resin Properties⁹

Base Resin: 06067 (HDPE)

<u>FILLER</u>	<u>I₂</u>	<u>I₁₀/I₂</u>	<u>DENSITY</u> <u>gm/cm³</u>	<u>RIGIDITY</u> <u>psi x 10⁴</u>
None	7.25	8.7	0.9644	17.5
5% Aluminum Silicate	7.49	8.4	0.9887	16.9
5% Zinc Oxide	5.73	9.5	0.9812	17.5
5% Talc	5.82	9.0	0.9876	18.5
10% Talc	5.15	9.4	1.0174	20.6
5% Aluminum	5.95	9.0	0.9884	17.4

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Table II

Filled HDPE⁵

<u>Filler</u>	<u>Source</u>	<u>Weight % Filler</u>	<u>Density gm/cm³</u>	<u>Room Temp. Izod Impact Ft.-Lbs./ inch</u>	<u>(Stiffness) Tange Modulus of Elastic PSI x 10⁴</u>
Thermax	R. T. Vanderbilt	33	1.11	0.60	17.1
Kosmos 60	United Carbon Co.	33	1.13	0.54	35.0
6 X Mica	Mineralite Corp.	33	1.19	1.24	18.3
Silene EF	P.P.G.	33	1.15	0.87	28.2
Asbestos 7F	Johns-Manville Co.	33	1.20	0.99	19.8
Sulfur	Freeport Sulfur	15	1.02	2.04	9.3
Micro Cell B	Johns-Manville Co.	33	1.14	0.48	20.5
	Dow Chemical	0	0.95	1.66	10.1
Super Cel	Johns-Manville Co.	33	1.15	0.97	29.7

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Table III
Filled LDPE²

<u>Filler</u>	<u>Source</u>	<u>Weight % Filler</u>	<u>Density gm/cm²</u>	<u>Stiffness PSI X 10⁴</u>	<u>Tensile Strength PSI.</u>	<u>Ultimate Elongation %</u>
Clay 33	LDPE D-55	0	0.92	19.6	1950	150
	Southern Clays, Inc.	40	1.22	41.0	1800	45

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Table IV

Filled HDPE⁴

<u>Weight % Filler</u>	<u>Type of Filler</u>	<u>Melt Index</u>	<u>Rigidity X 10⁴ PSI.</u>	<u>Tensile PSI.</u>	<u>Yield PSI.</u>	<u>% Elongation</u>	<u>Izod Impact</u>	<u>Tensile Impact</u>	<u>Density</u>	<u>Se Mod X 10¹¹</u>
0	CaCO ₃	6.17	14.8	2500	4420	640	0.60	1.44	0.963	13
25	"	5.11	20.9	3770	3770	0	0.37	0.82	1.15	15
37.5	"	4.56	31.1	3260	3260	0	0.22	1.09	1.28	14
50	"	2.23	35.0	CNA	CNA	CNA	CNA	CNA	1.44	C)
*70	CaSO ₃	0.07	61.1	5315	5315	5	1.94	17.4	17.4	24

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Table v⁴Talc Filled 06067 (HDPE)

<u>Property</u>	<u>0%</u>	<u>5%</u>	<u>10%</u>
Melt Index	7.25	5.82	5.15
Tensile	3225	-	-
Yield	4745	-	-
% Elongation	115	-	-
Rigidity	1.75×10^4	1.85×10^4	20.6×10^4
Density	0.9644	0.9876	1.0174
I ₁₀	62.76	52.67	48.52

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