EXHIBIT V

PART 22

45

Polyolefin Modification with EXACT™ Plastomers

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Introduction

The commercially available ethylene-based EXACT™ (1) polymers span a very wide range in molecular weights and comonomer contents. Many grades of polyethylene copolymers ranging from viscous high molecular weight resins to low molecular weight liquids, and from highly crystalline stiff materials to soft pliable adhesives, are possible. A new family of ethylene/alpha-olefin copolymers with low crystallinity has been developed. This type of product features uniform narrow composition distribution (CD) (2), in other words, a carefully regulated distribution of comonomers among the polymer chains. Unlike standard polyethylene copolymers, which often contain a highly crystalline HDPE end and an amorphous highly extractable tail, comonomer utilization by the single-site catalyst is much more uniform and efficient. Polymer crystallinity can be suppressed to that of the traditional elastomer region (density < 0.9 g/cm3). We call this type of resin "plastomer" to reflect its dual plastic and elastic features. Table I shows many of the characteristics of plastomers, reflecting both its plastic and elastomeric attributes. With a density range of 0.90 g/cm3 to 0.87 g/cm3, plastomers bridge the gap between very low density polyethylenes (VLDPE, 0.900 to 0.915 g/cm3) and traditional ethylene-propylene elastomers (EPR). In this paper we will first discuss the unique features of plastomers derived from their narrow composition distribution and second, on the miscibility of various types of plastomers with polyolefins, particularly polyethylenes and polypropylenes. Finally, several examples of plastomer applications are discussed in order to illustrate the uniqueness of this family of products in fulfilling specific requirements.

Composition Distribution

The composition distribution has a drastic impact on the melting behavior. For typical Ziegler catalyzed polyethylenes, the peak melting point is almost the same as its homopolymer regardless of its comonomer content. This is because this type of copolymers tend to have a very disparate CD with some molecules in a given material having high comonomer content while others are very low in comonomer content. The peak melting point of this type of product is mainly governed by the lower comonomer content, higher crystallinity segment. EXACT™ plastomers, on the contrary, show a linear dependency of melting temperature as a function of comonomer level. Figure 1 is

generated from actual DSC peak melting point measurement for many plastomer samples with known densities. Regardless of comonomer, melting points are linearly reduced with a corresponding decrease in density. The uniform comonomer distribution is also very effective in reducing the polymer's crystallinity. At 10 mole% or greater comonomer incorporation, crystallinity becomes so low that most of the plastomers become water-clear. These highly amorphous plastomers function well as impact modifiers for polyolefins, competing effectively against lower density and lower melt index ethylene-propylene elastomers.

Unlike many lower density polyolefins (density < 0.87), the plastomer pellets are non-sticky and free flowing. Dry blending is usually sufficient for molding or extrusion which can be done using the standard conditions for the base resin. Three grades of EXACT™ plastomers which are commonly used for polyolefin modifications are shown in Table 2. Contrasted with many of the pelletized, low density resins in the market today, the plastomers do not require any antiagglomerant agent (usually HDPE powder or talc), nor any processing aid such as stearates during manufacturing.

Miscibility

Many types of alpha-olefins can be used as comonomers for plastomers. The type and amount of comonomer incorporation influence the miscibility with polyolefins. A common miscibility predictor for non-polar polymers is the solubility parameter of Hildebrand and Scott (3). It is defined as the square root of the cohesive energy density. The concept behind the solubility parameter is that polymers with similar cohesive energy tend to disperse into a homogeneous mixture whereas a higher cohesive energy polymer not only requires more energy to disperse, but is also likely to return to its original state of existence after mixing with a lower cohesive energy polymer. Availability of the solubility data allows one to calculate the Flory-Huggins interaction parameter χ (4,5) which is a more refined predictor for polymer miscibility. The interaction parameter, for non-polar polymers, can be obtained by multiplying the square of the solubility parameter difference with the factor (V/RT), where V is the molar volume of the amorphous phase of the repeated unit, R is the gas constant, and T is the absolute temperature. Therefore, the χ value is always a positive number. A χ value of less than 0.002 is often reported in literature as the critical value to produce a miscible blend for 100,000 weight average molecular weight constituents at room temperature (6). Industrial experiences with thermoplastic polyolefins and elastomers suggested that polypropylene and EPR, though not miscible based on solubility considerations, form the so-called compatible blends. This type of blend is characterized by a fine dispersion of the rubbery component (approximately 1 micron in particle size) in a continuous matrix resin. Unlike the miscible blend, its properties are usually the weighted average of its constituent polymers (7, 8), the compatible blend has the advantage of producing synergistic properties, such as low temperature impact resistance in this case. It is, however, difficult to define a critical χ parameter for the compatible blends. Higher χ value blends can still be made compatible either by increasing the molecular weight of the dispersed phase or by a partial crosslinking of the dispersed phase to retard its

mobility. It is generally true that with the higher χ valued blends it is more likely for the dispersed particles to bloom to the surface with aging. A binary compatible blend is usually phase separated, but there are sufficient interlamellar tie molecules (9) or increased thickness of the interfacial region to allow for efficient stress transfer, so that in the case of a rubber modified blend, a tougher product is produced. A qualitative argument regarding the trend of the χ values for compatible blend is that the area fraction of the tie molecules, or the thickness of the interfacial region increases with decreasing χ value.

Solubility parameters obtained from many sources of reference are useless and most often lead to unreliable miscibility predictions. A method of calculating consistent solubility parameters for non-polar molecules by the well known group contribution method has been made recently available in a new computer software package from Pennsylvania State University (10). Solubility parameters for polypropylenes and polyethylenes, as well as many types of plastomers, are first calculated and then the solubility parameter differences among these polymer are tabulated as in Table 3. The solubility parameter difference calculated in Table 3 is next used by the same software program to calculate the Flory-Huggins interaction parameter χ, as shown in Table 4. All the calculations shown in Table 4 assume that the comonomer content for plastomers is 20 wt.%, and the propylene content in EPR is 35 wt.%. The variation of the interaction parameter χ with the plastomer type and composition is also easily obtained by the same program. In Figure 2 the composition dependence of the interaction parameter for EPR and five types of plastomers, i.e., ethylene-butene (E-B), ethylene-hexene (E-H), ethylene-octene (E-O), ethylene-decene (E-D), and 4-methylpentene (E-M) are also shown.

We can now draw several conclusions from Table 4 and Figure 2, assuming all the EPR and plastomers are of the same degree of polymerization. It is clearly shown in Figure 2 that none of the plastomers will be miscible with isotactic polypropylene (IPP) because of their χ values are much above the upper critical value for miscible blends, 0.002. In the case of EPR, only at very high propylene content does the EPR becomes miscible with IPP. The χ value increases slightly from a propylene comonomer to a butene comonomer, and then increases rapidly for the hexene comonomer. A maximum χ is reached when the comonomer is an octene. No further increase in χ value occurs when decene is used and its calculated χ value is the same as χ for the hexene comonomer. We may therefore conclude from this observation that the E-B type of plastomer will be more compatible with PP than the E-O type of plastomer. The ethylene-4-methylpentene plastomer, on the other hand, shows a χ value between propylene and butene, which makes it more compatible with PP than butene based plastomer, but at added comonomer cost.

Table 4 points to another interesting observation between EPR and plastomers. The calculated χ values for EPR/polyethylene blends are three times higher than the critical value for miscibility, therefore making it not miscible with polyethylene but only

compatible. On the contrary, all five types of plastomers show extremely small χ values below the critical miscibility value, and they should all be miscible with polyethylenes. The E-B plastomer is the material of choice because it is based on the lower cost comonomer. Table 4 also predicts that both EPR and plastomers are more compatible with impact polypropylene copolymer (ICP) than with IPP due to reduced χ value.

Modification of High Density Polyethylene

The following are selected examples to illustrate the utilization of the many plastomer attributes which bring about a better product. The major categories of EXACT™ modified products are shown in Table 5. It is interesting to note that plastomers and polyolefins can be used alternatively to modify each other. Plastomers can be used to improve the tear and impact of polyolefins, and similarly, polyolefins can modify the stiffness and melt strength of plastomers.

The miscibility of plastomers with high density polyethylene prompts us to develop clear plastomer/PE alloys to use as a potential PVC replacement in the healthcare market. Due to stringent environmental requirements for incineration at hospitals, a halogen-free product is desirable. One such application is for flexible vinyl based surgical disposable bags. Currently, the bag is produced via extrusion blow molding. In ideal circumstances, a drop-in product with minimum production hardware modification could be used. We have successfully developed such a material by dry blending 20 to 30 wt.% HDPE resin with a plastomer. A minor amount of HPDE serves to boost the melt viscosity necessary for extrusion blow molding. The produced bag is clear and flexible and, like the current vinyl one, it is easily collapsible inside a protective canister.

In a different application, about 20 to 30 wt.% of plastomer is tumble blended with HDPE pellets, and the dry blend is next used to produce a 4 mil overwrap blown film. The addition of a minor amount of plastomer greatly improves the machine direction (MD) tear strength of the film while maintaining a high film stiffness. Figure 3 shows the effect of plastomer addition. The MD tear strength increases with a corresponding decrease in secant modulus. Addition of 25 wt% plastomer will improve the MD tear strength to twice that of the HDPE control, and still maintaining much higher stiffness than an LLDPE film.

Modification of Linear Low Density Polyethylene

LLDPE is often used to produce plastic liner grade films. A typical construction consists of a LLDPE outer layer, a middle core layer, and a LLDPE/EPR lamination inner layer. Two roll of films are then laminated to produce the final product. Production rate is restricted by the line speed of the lamination operation to produce liners with required seal integrity. A laboratory study of heat seal strength as a function of temperature was conducted. As shown in Figure 4, the desirable plateau seal strength is achieved at a 50

% loading EPR/HD, but only 45% loading of plastomer is required to obtain the same seal strength. A second advantage of the plastomer blend is that the plateau seal strength is reached at much lower temperature than the EPR control blend, 80 °C versus 120 °C. Replacement of EPR/HD with EXACT™ plastomer therefore lowers the lamination temperature considerably and consequently allows for increase in line speed of the lamination operation. Table 6 displays a comparison for the properties of these two lamination films. No major difference in properties is detected, and an increase in productivity and reduced raw material cost is achieved.

In another application pelletized rubber is used to impact modify LLDPE which, in turn, is used to produce multilayer films Commercial rubber pellets, although excellent impact modifiers, have shortcomings. First, a dusting agent needs to be employed to prevent pellet agglomeration. Second, a small amount of processing aid such as an organic metal stearate is inevitably present. The pelletized rubber is produced by masticating bales of rubber in a high intensive mixing and the added heat history usually reflects as an increase in the yellowness index. Replacement of pelletized rubber with the colorless EXACT™ plastomer will eliminate above mentioned shortcomings.

A comparison of the effect of temperature and blend ratio on notched Izod impact values between the LLDPE/EPR and LLDPE/EXACT™ blends is shown in Table 7. There is virtually no difference in impact performance noticed from room temperature to - 40 °C, and at either 10 wt.% or 20 wt.%. An interesting phenomenon, commonly known as impact reversal is observed, namely, the notched Izod value measured at room temperature is found to be lower than that measured sub-zero temperature. This is usually found to be true during testing of a flexible test specimen, such as a LLDPE/EXACT™ blend. The ASTM Izod tester is designed in such a way that a flexible sample can be forced into a right angle fold to allow for the passage of the impact pendulum. As the test temperature is lowered, the stiffness of the test specimen increases, hence more energy is required to fold the sample, a higher Izod value is registered. The impact reversal phenomenon is more clearly shown in Figure 5. Plastomers with a constant density of 0.885 g/cm3 but with varying melt indexes from 1.5 to 9.7 are found to be equally effective in impact modification of LLDPE. The average Izod value is less than 6 ft-lbs/in at room temperature, but it gradually increased to 11 ft-lbs/in at - 18 °C, and finally it reaches 16 ft-lbs/in at - 40 °C.

An EXACT™ plastomer with its combined high melt index and low melting point make it easily dispersible in LLDPE. A comparison of the effect of twin screw versus single screw compounding on the impact properties was conducted. As shown in Figure 6, no distinguishable difference in impact between these two blends is noticed from room temperature to - 40 °C. Single screw compounding is sufficient, and no added advantage can be achieved by the utilization of a twin screw compounder. We have also noticed that when the amount of plastomer in a given blend is above 10%, even direct processing from a dry blend is often practiced.

Modification of Polypropylene

EXACT™ plastomers are used to modify all three commercial types of polypropylene. The simplest application is the addition of a small amount of plastomer to isotactic polypropylene (IPP) to upgrades its toughness. Another interesting application is impact modification of clarified random polypropylene copolymer (RCP) (11) as a lower cost polyethylene terephthalate (PET) replacement in both extrusion blow molding and injection molding applications. EXACT™ plastomer is an ideal modifier to improve the drop resistance of a clarified RCP with minimum increase in haze. This is demonstrated in Table 8, that addition of 10% of plastomer exhibits the same haze level as unmodified clarified RCP. When 20% plastomer is added, the ductile-brittle temperature is lowered to below 20 °F with only a slight haze increase. Table 8 also shows that no difference in haze is noticed between test plaques injection molded from dry blend and melt blend.

The recent trend toward reduction of the number of types of automotive plastics for recycling purposes accelerated the development of thermoplastic polyolefins (TPO). EXACT™ plastomers can be used to tailor the cold temperature impact of impact polypropylene copolymer to produce TPOs. A high speed instrumented falling weight impact experiment was conducted to compare the effectiveness of EXACT™ plastomers with several commercial polypropylene modifiers. The instrument used was a Ceast Fractovis fitted with a 20 mm hemispherical striker, and a 40 mm anvil span for the sample holder. Test was conducted a 4 m/second impact speed, and from room temperature to - 40 °C. Data collected were maximum energy, which represents the onset of penetration of the test specimen by the high speed striker, and total energy which corresponds to the complete penetration of the test specimen. A ductility index can be calculated by first subtracting the maximum energy from the total energy and next dividing the difference with the total energy. For an ideal ductile material, the total energy is assumed to be twice of that of the maximum energy. Hence, the ductility index is 0.5, and similarly for a brittle material, the sample shatters immediately after impact, its ductility index is zero. The ICP control tested at room temperature absorbed a maximum energy of 28 ft-lbs, and a maximum energy of 45 ft-lbs which produces a ductility index of 0.38. At - 40 °C, its ductility index was reduced to only 0.20.

Figure 7 shows the impact enhancement of incorporating 30 % EXACT™ plastomer to ICP. In the same figure, the impact data for 30 % EPR, EPR/HD, and a commercial grade of broad composition distribution (BCD) plastomer, are also shown. The impact reversal phenomenon is again observed during testing, and both maximum and total energies increase with lowering of test temperature. All four types of modifiers show comparable maximum energies, but the BCD plastomer modified ICP shows much lower total energy at -40 °C test temperature. The ductility index as a function of temperature is next shown in Figure 8. Again the BCD plastomer modified ICP shows a rapid decline in ductility index with lowering in test temperature, whereas only a slight drop in ductility index with lowering temperature is observed for the other samples. At - 40°C,

the ductility index of BCD plastomer modified ICP is only slightly higher than the unmodified ICP, 0.26 versus 0.2, in compared with a 0.45 ductility index for EXACT™ modified ICP.

Finally, the EXACT™ modified ICP blend shows less propensity to produce stress whitening than other three modifiers mentioned above. As shown in Figure 9, stress whitening diminishes with the increased amount of plastomer and addition of all other three modifiers show no stress whitening improvement. In the case of modification of PP, all four modifiers induce generation of stress whitening. EXACT™ plastomer is able to distinguish itself from EPR, EPR/HD, and BCD plastomer in that it generate a lot less stress whitening.

Conclusion

The above examples illustrate the versatility of EXACT™ plastomers for the modification of polyolefins. A variety of other applications are under development. Wire and cable compounds are being developed in order to take advantage of the high filler loading capacity of plastomers, and color concentrate with high pigment loading can now be achieved with the replacement of polyolefins by the plastomer. Their excellent filler acceptance makes them prime candidates for base resins in all types of additive masterbatches. These new color concentrates and masterbatches will be compatible with polypropylene and polyethylene as illustrated above. The excellent compatibility of plastomers with polyolefins can also serve as a dual compatibilizer/impact modifier to upgrade the performance of comingled recycled plastics.

Obviously, EXACT™ plastomers are expanding the meaning to the term "modification". We have seen not only improved impact performance, critical for so many PP and PE applications, but also improved clarity, seal strength, toughness, and stress whitening performance. Additionally, the lower extractables of the narrow composition distribution plastomers allow for polyolefin modification with FDA compliance. The overall performance is best viewed by using a star plot for selected properties. It is shown in Figure 10 to describe the key property profiles of EXACT™ modified ICP versus EPR modified ICP at 30% modifier level. The applications are extensive and unique. Many will require unique polymers. Fortunately, Exxon Chemical's EXACT™ polymers plant in Baton Rouge, LA, has the versatility to produce a wide variety of ethylene-based products, and we are committed to work closely with our customers to identify the plastomer that best suits their unique modification need.

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Table 1 Characteristics of EXACT™ Plastomer

Plastic Attribute	*Elastomer Attribute*
Free Flowing Pellets	Low Melting Point
Dry Blending	Low Crystallinity
Low Odor	Colorless
No Dusting Agent	Filler Acceptance
Density	Oil Acceptance
Melt Index	ESCR
Low Extractables	Adhesive

Figure 1 Melting Points of EXACT™ Plastomers

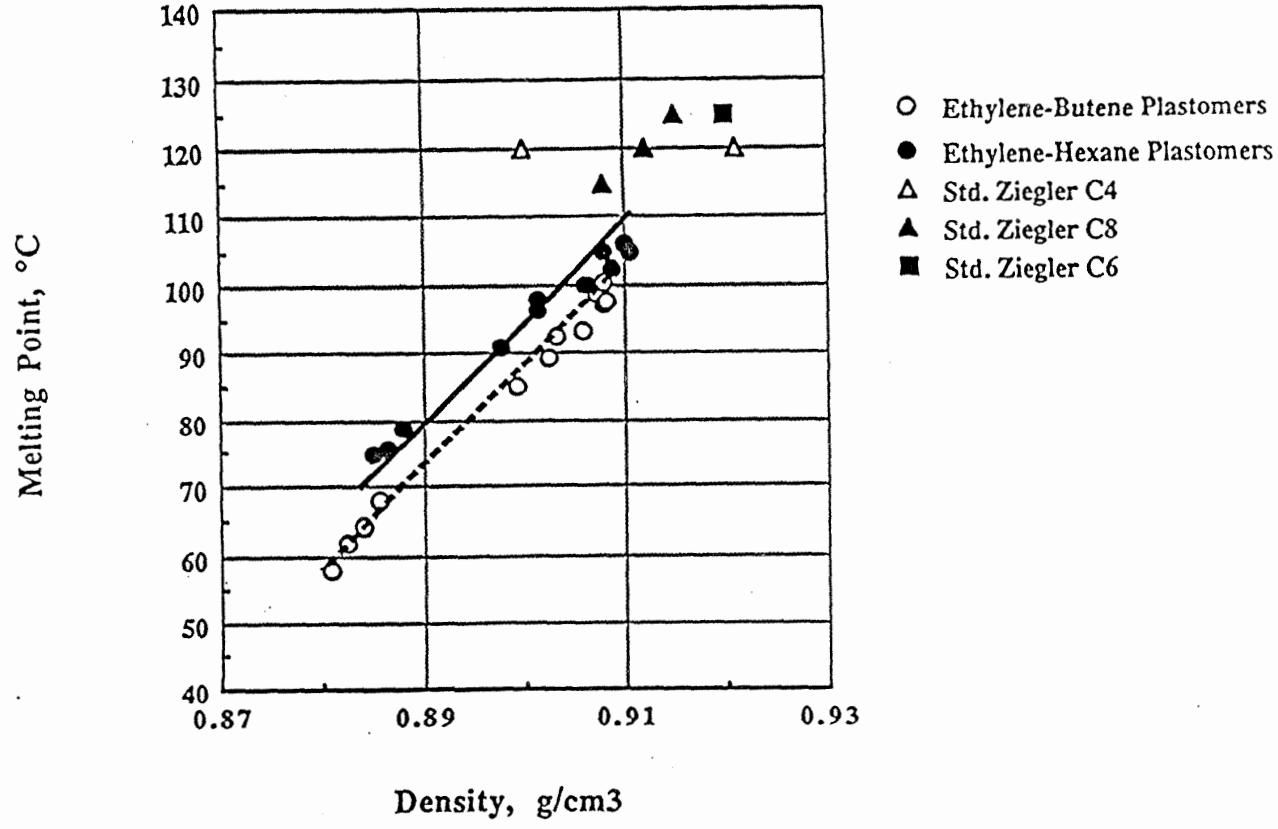

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Table 2 EXACT™ Polyolefin Modifiers

Grade	Comonomer	Density	Melt Index	Color	Application
EXACT™ 4033	Butene	0.88	0.8	Colorless	Premium Low Temp. Impact
EXACT™ 4041	Butene	0.88	3	Colorless	High Flow/High Impact
EXACT™ 4042	Butene	0.9	0.9	Colorless	FDA*/High Impact
Pelletized EPR	Propylene	0.86	0.3	20 Max YI	FDA**/High Impact
HDPE Extended EPR	Propylene	0.885	0.4	20 Max. YI	Dry Blend/High Impact

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* *21CFR177.1520 (c)3.1 or 3.4*

** *21CFR177.1520(a), (b), (c)3.4 and 3.5*

Table 3 Solubility Parameter Differences, (cal/cm3) ^ 0.5

Plastomer (Solubility Parameter)		Polyolefin (Soulbility Parameter)			
	IPP (7.43)	RCP (7.44)	ICP (7.49)	LLDPE (7.97)	HDPE (7.99)
Ethylene-Propylene Elastomer (7.79)	0.36	0.35	0.3	0.18	0.2
Ethylene-Butene Plastomer (7.91)	0.48	0.47	0.42	0.06	0.08
Ethylene-Hexene Plastomer (7.94)	0.51	0.5	0.45	0.03	0.05
Ethylene-Octene Plastomer (7.96)	0.53	0.52	0.47	0.01	0.03
Ethylene-Decene Plastomer (7.94)	0.51	0.5	0.45	0.03	0.05
Ethylene-4-Methylpentene Plastomer (7.90)	0.47	0.46	0.41	0.07	0.09

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Note: Calculations are based on comonomer content of 35 wt.% for ethylene-propylene elastomer, and 20 wt.% for plastomers

Table 4 Flory-Huggins Interaction Parameter

Plastomer (Solubility Parameter)		Polyolefin (Solubility Parameter)			
	IPP (7.43)	RCP (7.44)	ICP (7.49)	LLDPE (7.97)	HDPE (7.99)
Ethylene-Propylene Elastomer (7.79)	0.023	0.022	0.016	0.006	0.007
Ethylene-Butene Plastomer (7.91)	0.039	0.04	0.032	< 0.002	< 0.002
Ethylene-Hexene Plastomer (7.94)	0.044	0.042	0.034	< 0.002	< 0.002
Ethylene-Octene Plastomer (7.96)	0.047	0.045	0.037	< 0.002	< 0.002
Ethylene-Decene Plastomer (7.94)	0.044	0.042	0.034	< 0.002	< 0.002
Ethylene-4-Methylpentene Plastomer (7.90)	0.038	0.036	0.03	< 0.002	< 0.002

Note: Calculations are based on comonomer content of 35 wt.% for ethylene-propylene elastomer, and 20 wt.% for plastomers

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Figure 2 Flory-Huggins Interaction Parameter Polypropylene/Plastomer Blends

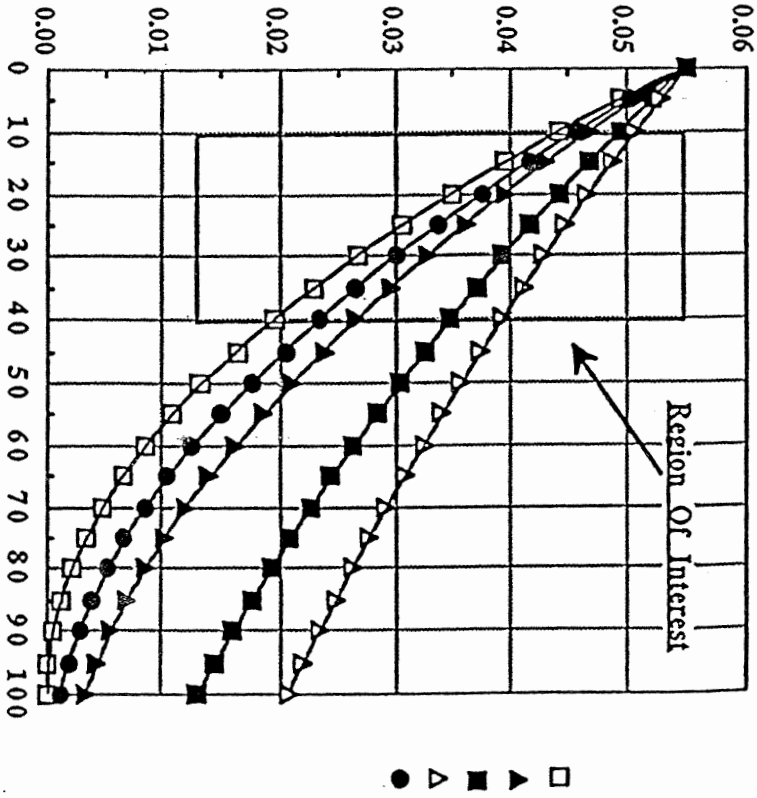

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Table 5 EXACT™ Polymer Modifier Applications

Type of Product	Major Component	Application	Process	EXACT™ Attributes
HDPE/EXACT™	HDPE EXACT™	Overwrap film Surgical disposable bags	Blown film Extrusion blow molding	TD tear strength Clear, flexible vinyl replacement
LLDPE/EXACT™	LLDPE EXACT™	Heat seal layer Shower curtain	Coex blown/cast film Cast film	Brodened heat seal range Clear, flexible vinyl replacement
RCP/EXACT™	RCP* EXACT™	Freezer resistance food server IV Bag	Injection molding Cast film	Low temp. impact, and low haze Impact, tear, clearity
EVA/EXACT™	EXACT™	Automotive bumper energy absorber	Injection molding	Filler acceptance, elongation
IPP/EXACT™	IPP	Wide-spec upgrade	Injection molding	Impact/elongation improvement
ICP/EXACT™	ICP	Automotive TPO parts	Injection molding	Low temp. impact, and ductility

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Figure 3 EXACT™/HPDE Blends

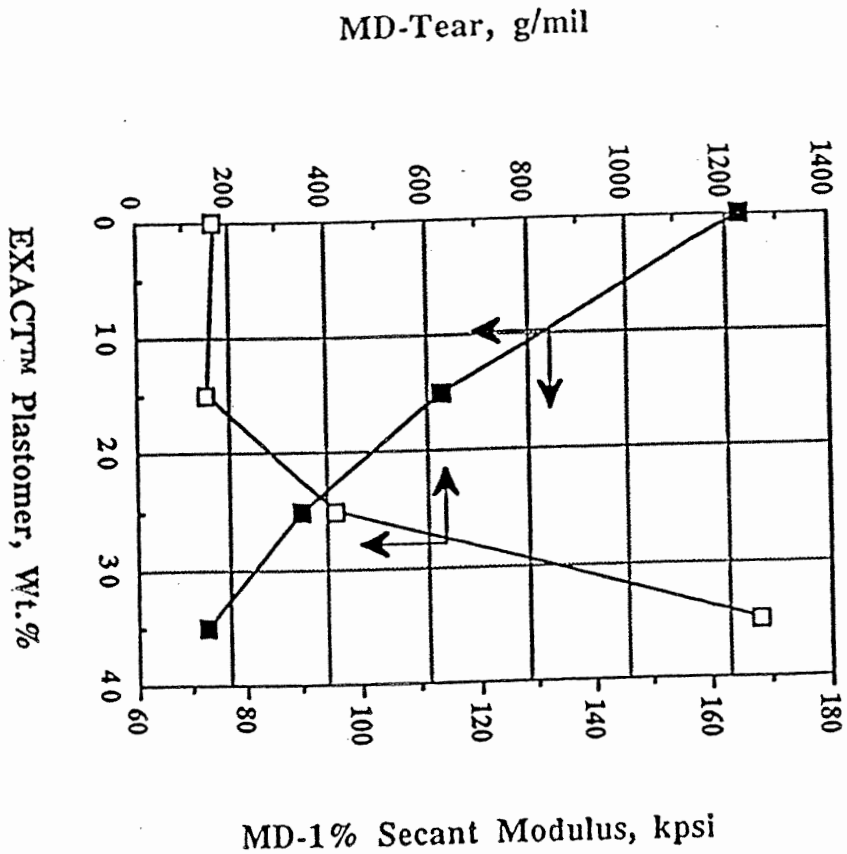
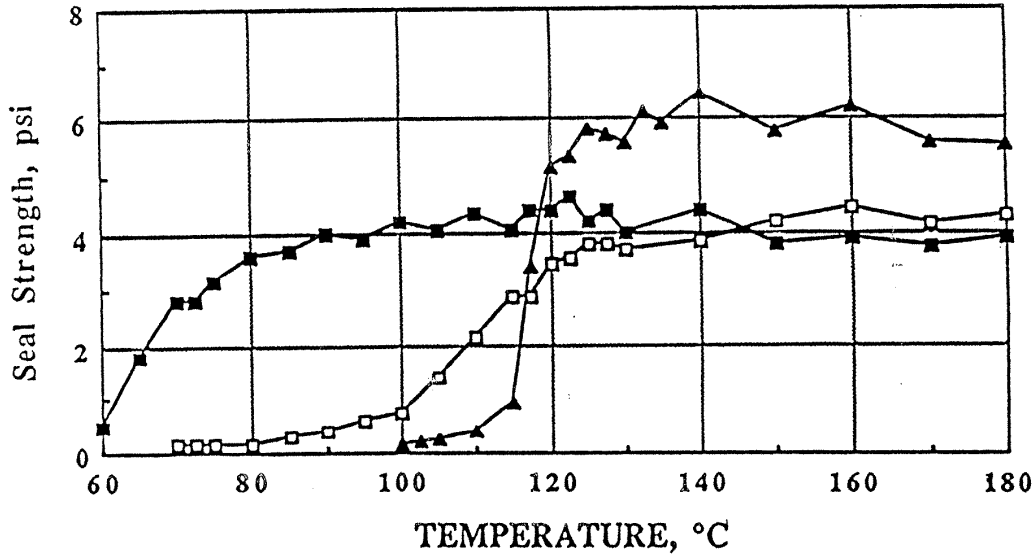


Figure 4 Heat Seal Behavior for EXACT™ Plastomer Modified LLDPE



Theller Model EB Heatsealer
1.3 second dwell time
Heat seal pressure 50 N/cm3 (11.2 psi)
Film thickness 3 mils

- LLDPE (Escorene LL 3001)
- 50% EPR (0.3MI/0.860 density)
- 45% EXACT™ Plastomer (3 MI/0.88 density)

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Table 6 Film Properties of EXACT™/LLDPE Blend

Property	LLDPE	50% EPR	45% EXACT™ 4041
Tensile Stress, psi MD/TD	6840/6100	5140/4460	5820/5160
Elmendorf Tear, g/mil MD/TD	400/590	150/340	230/320
Ultimate Elong., % MD/TD	810/820	680/860	800/820
1% Secant Modulus. Kpsi MD/TD	35/42	15/16	15/15
Dart Drop @ RT, g/mil	193	230	252

Film Thickness: 3 mils

Legend:

EPR: 0.3 MI/0.86 density
EXACT™: 3 MI/0.88 density
LLDPE: Escorene LL 3001

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Table 7 Notched Izod Comparison

(Ft-lbs/inch)

Type of Blend	23 °C	0 °C	- 40 °C
80/20 LLDPE/EXACT™	6.4 (NB)	6.4 (NB)	14.7 (NB)
80/20 LLDPE/EPR	4.7 (NB)	6.6 (NB)	14.9 (NB)
90/10 LLDPE/EXACT™	7.2 (NB)	7.2 (NB)	15.8 (NB)
90/10 LLDPE/EPR	5.4 (NB)	8.2 (NB)	17.0 (NB)

Note: 1/8" test specimen

Note: NB denotes no break

Legend:

LLDPE : Escorene LL-1002

EXACT™ : 1.5 MI/0.886 density, ethylene-butene copolymer

EPR: 0.3MI/0.860 density, ethylene-propylene copolymer

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Figure 5 Impact Profile for EXACT™ Plastomer Modified LLDPE

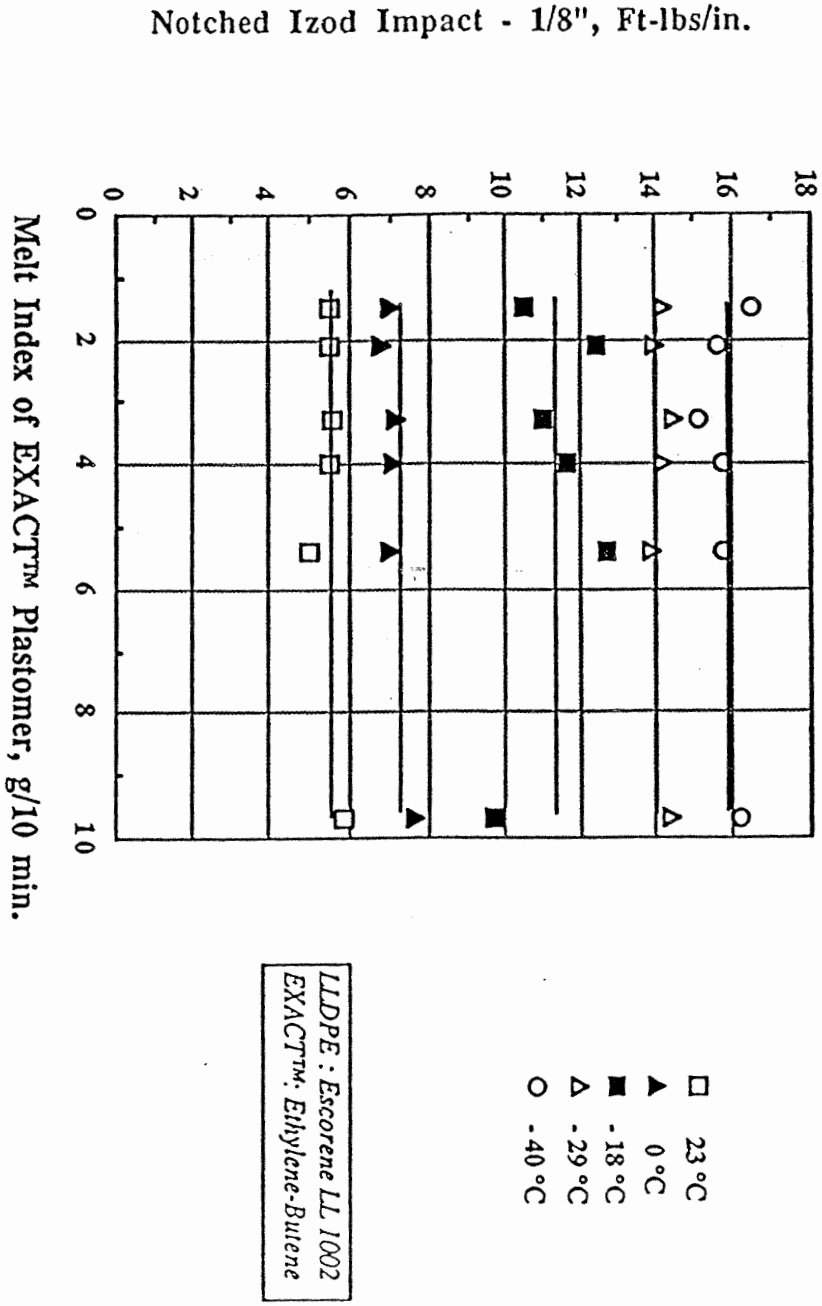

Figure 6 EXACT™/LLDPE Blends
Twin Screw vs. Single Screw

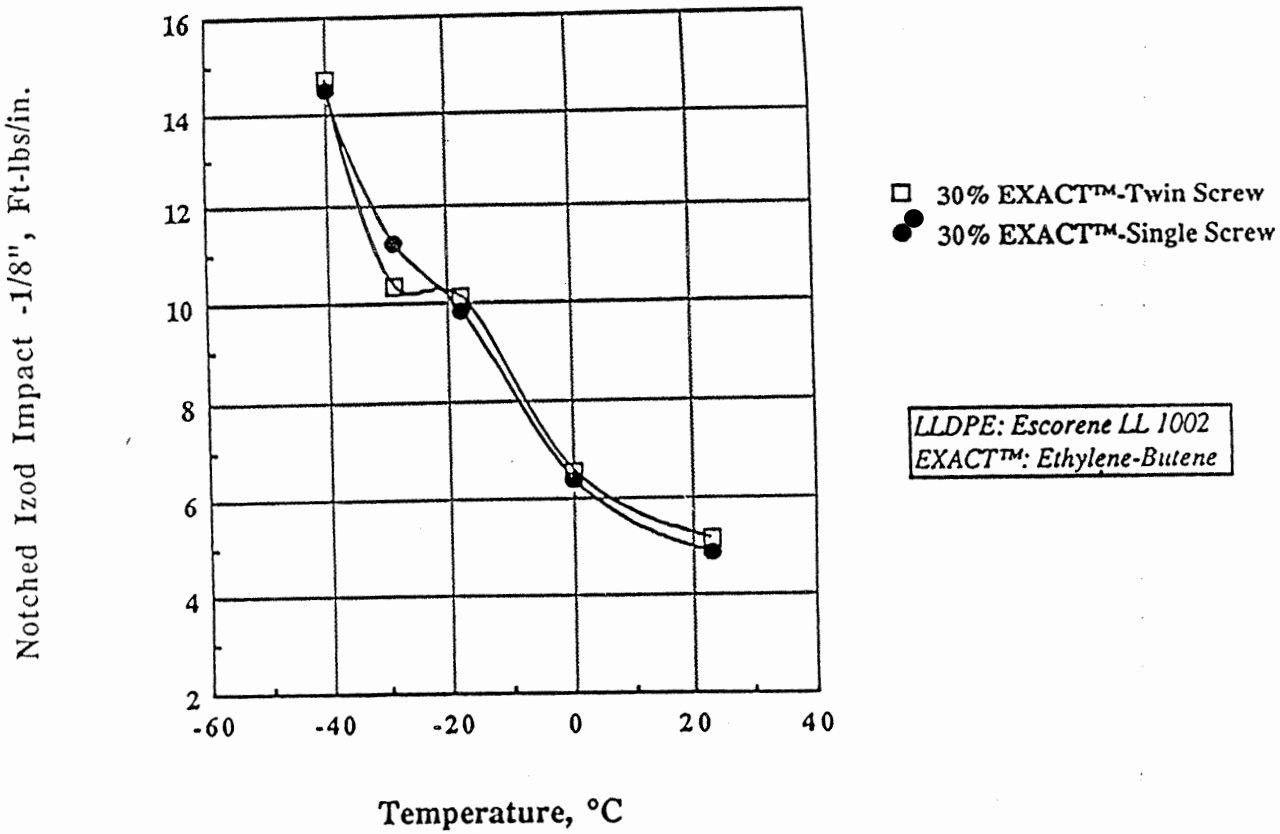

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Table 8 Effect of EXACT™ Addition on Haze

Copmposition	Dry Blend	Melt Blend
Clarified RCP	26.70%	26.70%
6% EXACT™	26.70%	26.10%
8% EXACT™	26.60%	26.60%
10% EXACT™	27.10%	28.30%
20% EXACT™	27.50%	28.50%

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Sample Thickness : 67 mils

Legend:
Clarified RCP: 12 MFR, 3% ethylene
EXACT™ : 3.4 MI/0.90 density, ethylene-butene copolymer

Figure 7 Instrumented Impact for 30/70 Impact Modifier/ICP Blends

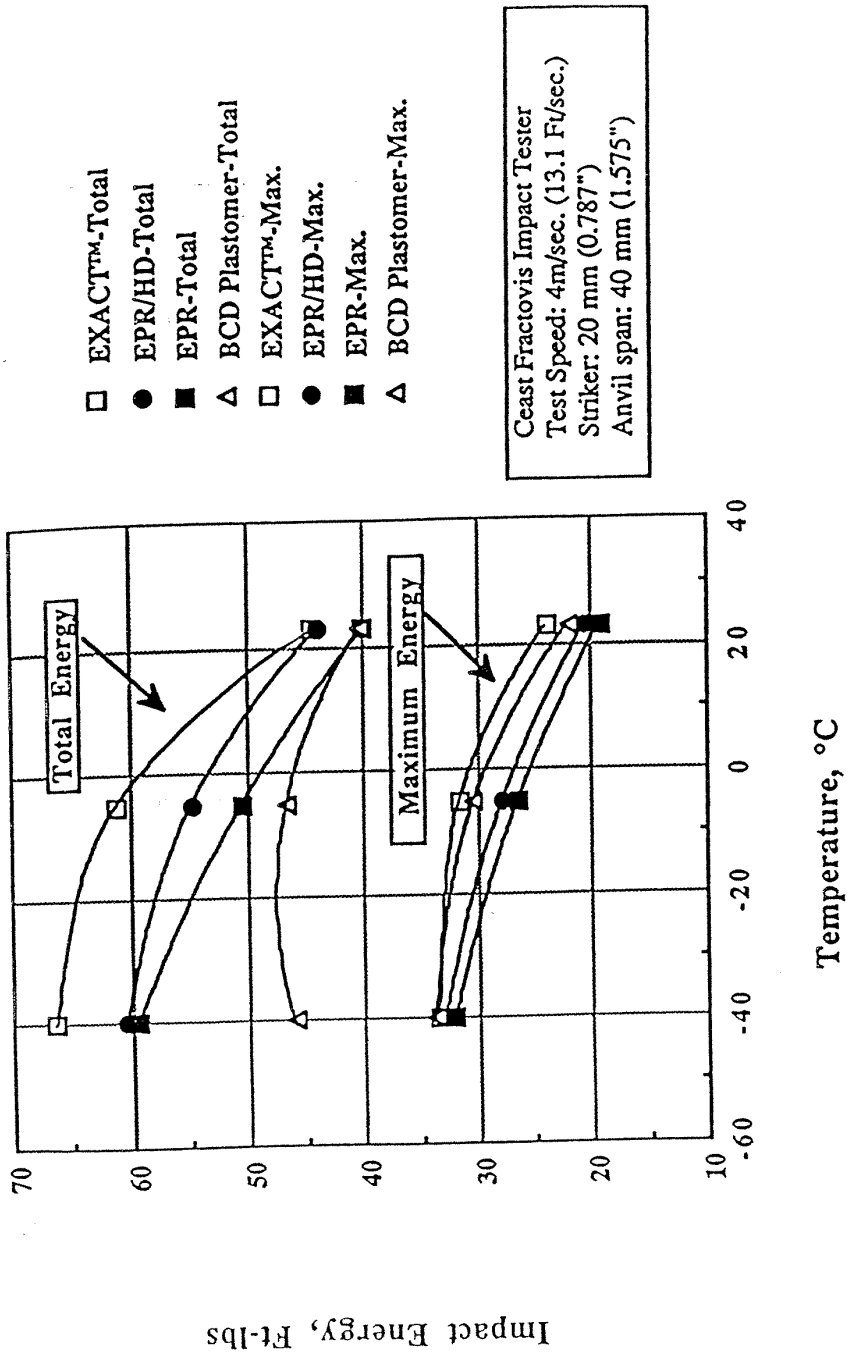

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Figure 8 Ductility Index for 30/70 Impact Modifier/ICP Blends

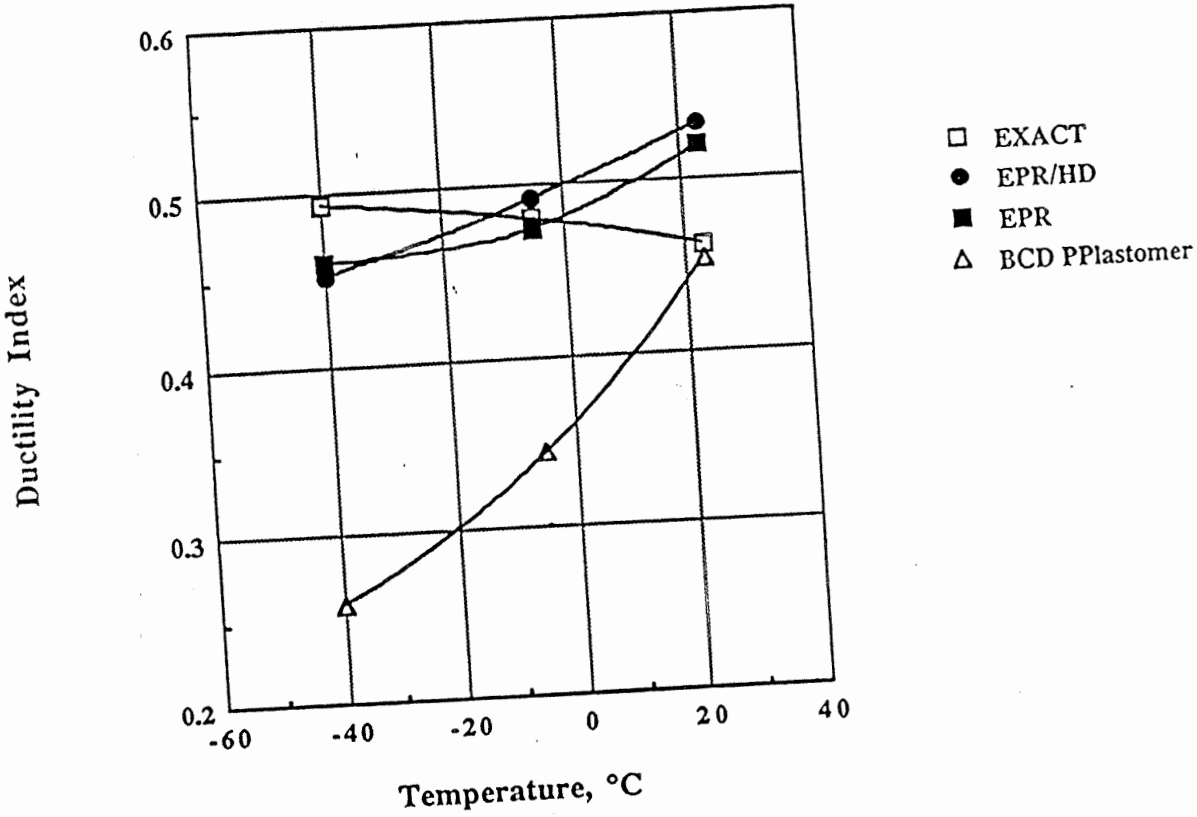

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Figure 9 Stress Whitening Index for Impact Modified Polypropylene Blends

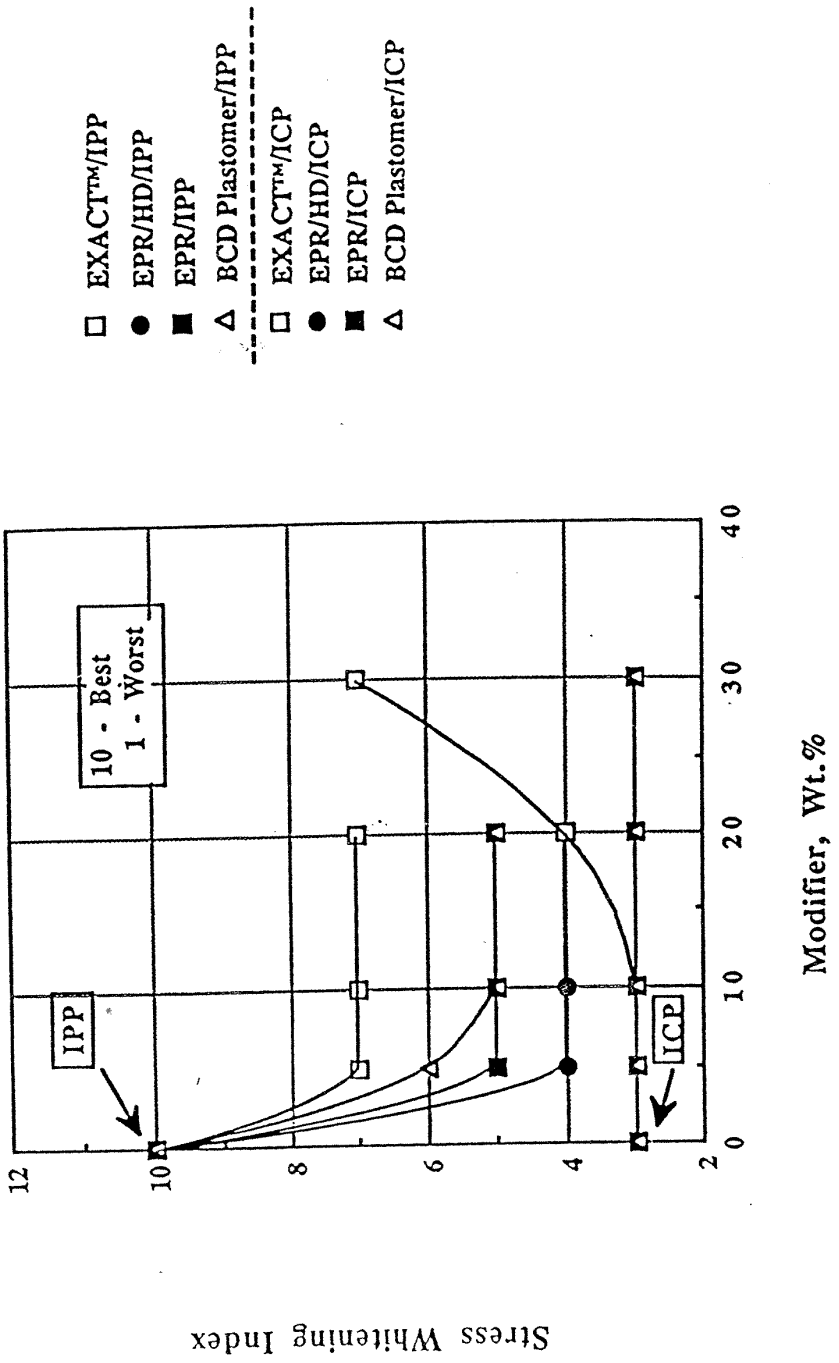
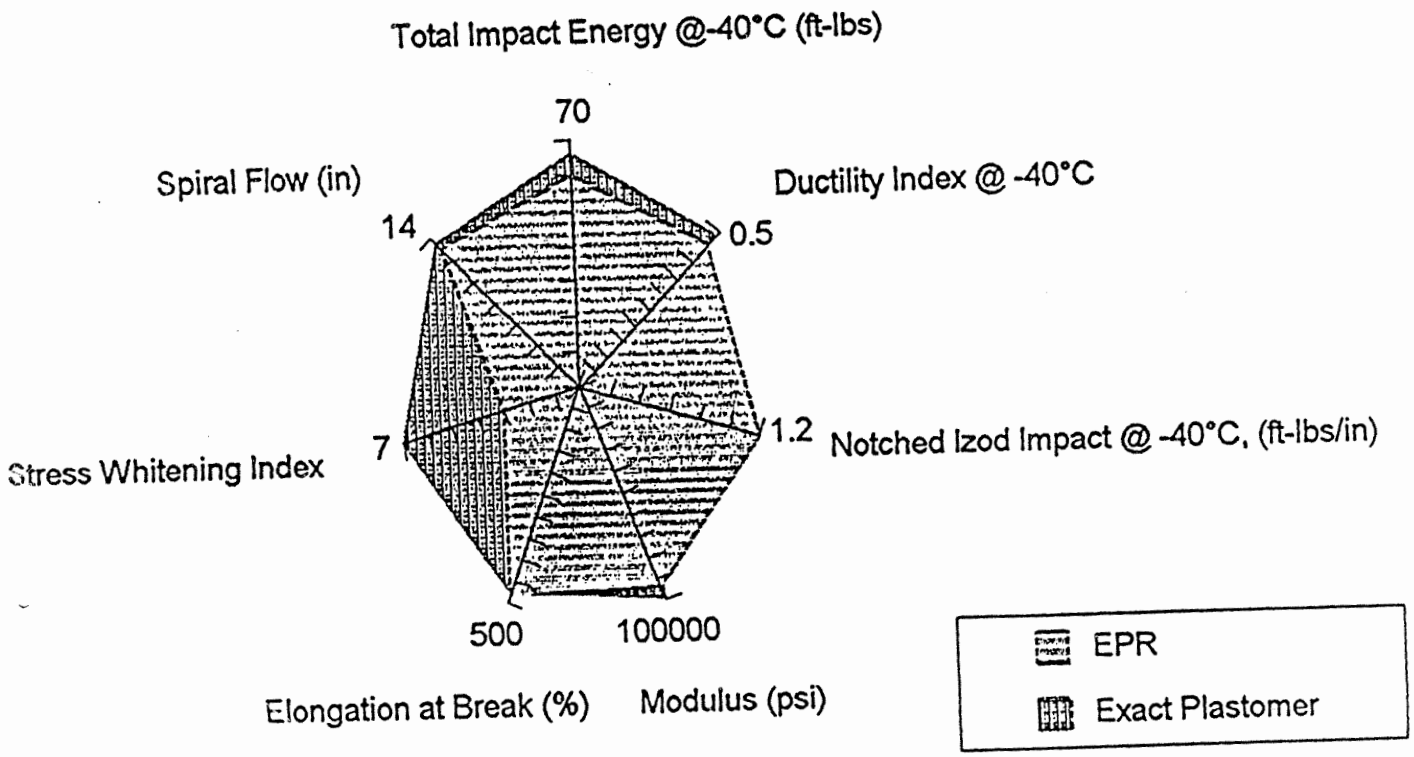


Figure 10 Star Plot for 30/70 EXACT-4/ICP Blend



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FUTURE TRENDS IN POLYOLEFINS TECHNOLOGY

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FUTURE TRENDS IN POLYOLEFINS TECHNOLOGY

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One way to get an idea of what the future might hold is to first take a look at the past. That's what I did in considering the topic for this meeting, namely... Future Trends in Polyolefins Technology... which also influences trends in the industry in general. In doing so, I noticed an interesting pattern about historic developments in this fascinating field.

The pattern concerns time. When reviewing past developments in polyolefin technology, I asked myself which, historically, were the most significant and when did they take place. These answers came to mind:

POLYOLEFINS HALL OF FAME

20 Year Milestones:

1930s Development of high-pressure process for PE

1950s Discovery of stereoregular polyolefins

1970s Development of low-pressure gas phase technology for linear polyolefins

1990s Commercialization of metallocene single site catalysts

• First, the development by ICI in the 1930s of its high-pressure process for making polyethylene. If there were such a thing as a polyolefins hall of fame, that pioneering technology would most certainly occupy a place of honor since it gave us the first commercial process for making this versatile plastic.

• Next in the 1950s came the discovery of stereoregular polyolefins and the incredibly rapid development of catalysts and processes to commercialize crystalline, isotactic polypropylene.

• It took me a bit longer to come up with the next entry but I think the development and licensing of the low pressure, gas phase Unipol technology for making linear polyolefins is a candidate for honors in the 1970s.

It's interesting to note these three milestones came at approximately 20-year intervals. Now, here we are in the 1990s... and, I believe that we have another once-in-every-20-year-milestone in polyolefin technology: namely, the commercial introduction of metallocene single site catalysis.

A few companies, including mine, have been working on single site catalysts for 10 or more years. But the first operating units using this new technology have come onstream only in the past year or so. That means single site catalysts are definitely products of the 1990s and beyond. As such, they keep intact a 20-year progression dating back some 60 years, and inevitably color any discussion of future trends in polyolefin technology.

So important is the single site catalysis' impact upon polyolefin technology that it's likely to drive four major trends in the industry itself.

SSC DRIVING INDUSTRY TRENDS

1. Delineation between SSC licensors and licensees
2. Proliferation of alliances and partnerships
3. Upward changes in profitability and growth
4. Expansion of SSC beyond polyolefins to other polymers

• Industry trend number one: We'll see an industry divided into two populations. One group will contain companies who've developed their own single site catalyst systems and patented technology. The other will contain those who license these capabilities. There will be few companies in the former category... and many in the latter. The reason for this is simple. Developing one's own single site technology will take huge amounts of time and money. This is most assuredly not a field in which you go out, hire a couple of chemists, and tell them to whip up a batch of new catalysts.

Entry in the field requires an extensive science-based understanding of catalysis that can only be built up over time and with such significant costs that only the largest companies will be able to afford the effort.

Licensing — always important in the industry — will become even more financially attractive for both

populations with single site catalysts, and the industry's traditional reliance on a few companies selling technology to the many will intensify.

The seller will see licensing income contribute significantly to profitability, and the buyer will have economical access to valuable new technology without a huge investment in time and resources.

• Industry trend number two: There will likely be more alliances and partnerships between companies with complementary science and research talents. Alliances, such as my company's with Mitsui and the relationships we've formed with customers, will proliferate. They reflect the reality that few companies who choose to "self-develop" can go it alone in this high-stakes endeavor and will need the complementary talents of others to commercialize the technology effectively and quickly.

• Industry trend number three: Profitability and growth will change significantly. The capabilities and potential of the new catalysts could well help change our industry's financial plight. These new catalysts will produce new polymers that will find more and more opportunities to penetrate new applications and replace higher cost polymers, thus impacting future demand growth.

• Industry trend number four: Single site catalysis will extend into polymers other than those made from olefins. Thus we'll be seeing catalyst changes in other polymers systems as well and there could develop real technology and cost synergy for doing so.

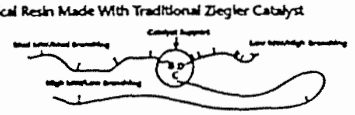
Now, let's take a look at the chemistry of single site catalysis... then how that chemistry is influencing trends in the industry's technology.

SINGLE SITE CATALYSTS VERSUS CONVENTIONAL CATALYSTS

EXACT Resin Made With EXXPOL Catalyst



Typical Resin Made With Traditional Ziegler Catalyst



All catalysts have reactive sites enabling them to perform their function — linking monomers into polymers.

Conventional catalysts have many sites of different activity levels located on their surface. The difference between the activity of each site introduces variations in the polymers they produce.

These new catalysts also have many reactive sites... but each reactive site is identical to all the others. This does several things:

• It does away with variability in the polymerization reaction.

• It allows a tailor-made structuring of the polymer.

• It permits an unsurpassed control of molecular weight distribution.

• It allows us to incorporate with great precision comonomers and termonomers and to make polymers with narrow composition distribution.

These catalysts produce incredibly consistent resins. We have seen combinations of properties not otherwise available, properties such as low heat-sealing temperatures, low haze, high clarity, impact resistance, elasticity and potentially — improved recyclability.

In short, these catalysts allow us to make a resin exactly right for specific end-uses — fulfilling a clear and growing need of polyolefin customers. This simple statement has implications so huge I want to say it again: these new catalysts allow us to tailor a resin exactly right for specific end uses.

Phrases such as "new and improved" are often worked to death. But our evidence is strong that single site catalysis makes possible "new and improved" polyolefins that eventually may replace more expensive materials such as engineered thermoplastics and high-performing elastomers or less environmentally benign materials like plasticized PVC and thermoset rubbers.

These new catalysts are here now... not on the horizon. My company has been operating a high-pressure polymerization line using single site catalysts at our Baton Rouge plastics plant since June of 1991. This unit produces 15,000 tons of ethylene polymers a year. It delivers a wide range of resins and supports an aggressive, worldwide product-development effort.

In addition, we are working with Mitsui Petrochemical Company to develop our own gas phase process application incorporating this new catalyst technology.

So much for some the expected technology driven industry trends and the "commercial." Now, let's see what trends single site catalysts might bring about in our polyolefin technology.

SSC DRIVING TECHNOLOGY TRENDS

1. Catalyst systems
2. Monomers & comonomers
3. Polymerization processes
4. Polymer products
5. Processing/converting
6. Markets for SSC-based products
7. Properties and attributes

I see polyolefin technology as a chain with seven links.

Link number one... the catalyst systems themselves.

Link number two... the monomers and comonomers we start with.

Link number three... the processes we use to polymerize these monomers and comonomers.

Link number four... the polymers these processes produce.

Link number five... the challenges facing those who process these polymers in resin form.

Link number six... the uses or applications to which these resins are put.

Link number seven... the properties of the end use products.

Single site catalysis is likely to affect each of those links.

CATALYST SYSTEMS

- Highly active catalysts
- Less residue, higher purity polymers
- Wide catalyst chemistry
- May help ZN catalysts work better

Let's look at link number one... catalyst systems. Single site catalysts are high-activity catalysts. That means they make many pounds of polymer per pound of catalyst. In addition to lessening the amount of catalyst needed to do the job — which can be an important economic consideration, there's a smaller amount of catalyst residue in the finished product.

These catalyst systems offer many different metallocene cations and anionic coactivator systems — each gives unique polymers and collectively they offer broad polymer design capability.

Then there's the area of mixed catalyst systems. We don't foresee the disappearance of Ziegler-Natta catalysts. They do many things well and people in the industry are comfortable working with these old friends. We can, however, envision a future in which users of these conventional catalysts will possibly be able to use the new family of catalysts to tweak their systems, to make them work better.

In addition, it's likely a single reactor will use conventional catalysts and single site catalysts at different times. And inevitably, there even may be some intermixing of the two catalyst systems in the polymerization process.

MONOMERS AND COMONOMERS

- Conventional catalysts polymerize large monomers with difficulty
- SSC copolymerize large and small monomers readily
- Will give broader range of materials

Link number two consists of the monomers and comonomers we're out to polymerize. Monomers come in different sizes. Some — like ethylene and propylene — are small. Others — like the higher alpha olefins and dienes — are big. Conventional catalysts have a classic limitation: They work well with little monomers but their reactivity drops way off when dealing with their larger brethren. The result is less copolymerization.

We can see where single site catalysts will be tailored to polymerize monomers of varying sizes. This could permit us to combine large and small monomers into the same polymers, thereby capturing the attributes and characteristics of both, thus likely giving us a broader range of starting materials to play with in developing new polymers and better products.

POLYMERIZATION PROCESSES

- SSC retrofittable into single and series reactors
- May allow switching from one polymer to another
- May produce new plastic/elastomer polymers
- May allow adding functionality for better properties

Link number three is processes. One approach we might see in tailoring polymers to meet customer needs is using a single site catalyst in a series of reactors... perhaps adding another monomer or another reagent as we go along. Or perhaps we'll see single reactors using a mix of SSC catalysts. We, and perhaps others, are looking into both methods.

Then there's the question of retrofitting... taking hardware already up and running with conventional systems and finding ways to introduce the new catalysts. With the vast amounts of installed capacity already running on conventional catalysts, this is an area of great interest.

Now here's another interesting thought on the impact of single site catalysis on processes: We all know that companies build different polyolefin plants to make different products like polyethylene, polypropylene and ethylene-propylene rubber. With the ability of single site catalysts to polymerize monomers of varying sizes and types, a single plant might be able to switch from one polymer to another with a minimum of fuss. Imagine ONE plant making polyethylene this month... polypropylene next month... and ethylene-propylene rubber the month after that.

This is one dimension of what we call blurring. Plastics with rubbery characteristics and plastic-like rubbers illustrate a second dimension of blurring. The interchangeability of processes and products demonstrate the potential of single site catalysts to make polymers do what needs to be done to meet customer needs.

Another likely area of continuing inquiry concerns functional polymers. Over the years, a good deal of work has been done on ways to polymerize monomers containing oxygen or nitrogen, two elements highly poisonous to conventional catalysts. The goal: to find ways of incorporating oxygen and nitrogen into the polymer, thus adding properties not otherwise available, without impairing the effectiveness of the catalyst. There's a real hope that single site catalysts may overcome this problem.

PROCESSING/CONVERTING

- Narrow MWD has many pluses, one challenge
- Flow characteristics being addressed
- Problem will be solved economically

Link number five in our chain has to do with fabricators who process resins into products... the customers of companies such as mine. Here again, we have opportunities but with a challenge: Polymers made with single site catalysts have narrow molecular weight distributions and no long-chain branches. This makes them more viscous and, consequently, often require more time and energy when going through a resin processor's equipment.

Though not a new problem — flow characteristics were a problem when the Unipol technology first came out — it's one that will get a lot of future attention from single site catalyst users. The use of improved processing aids or multiple reactors and multiple catalyst systems mentioned earlier are possible ways this problem will be economically solved.

We have developed a good deal of knowledge about markets — which I have made the sixth link in our technology chain. This knowledge results from an energetic product development effort undertaken with selected customers and with a growing list of new products. At present, a significant portion of the capacity of our Baton Rouge unit is devoted to developing new products.

To evaluate the properties of these resins, our technical service and support people are working with partners among customers in North America, Europe and Japan. These efforts are focusing on packaging film, electrical, automotive and medical devices, and textiles and adhesives.

MARKETS: Film Packaging

- Superior flexibility, clarity and cleanliness
- Disappearance of problems caused by inconsistent resins
- Tougher package integrity
- Downgaugeability possible

Here are some things this program has told us about just one application area — film packaging:

- We see customers reporting superior flexibility, clarity and cleanliness in molded and extruded health-care articles.

- Processing problems resulting from inconsistent resins virtually disappear.

- In film packaging we are in the process of commercializing polymers made with single site catalysts for selected high-value packaging applications. Our customer "partners" already have seen enhanced performance in film sealability, toughness, optics and cleanliness.

- Down gaugeability is another important attribute. The ability to use a thinner, tougher packaging film benefits producers — who need less material — and, when disposed of, has less bulk, an important waste-disposal consideration.

PROPERTIES AND ATTRIBUTES

- Uniformity and consistency
- Strength and elasticity
- Clarity
- Better organoleptics
- Thermal stability

That brings us to our seventh and last link in the polyolefin technology chain... the end-use attributes these catalysts will impart to the products made with our polymers.

Product attribute number one: uniformity and consistency. Single site catalysts give you an unrivaled control of the molecular weight distribution and composition distribution in the resulting polymers. We've unequivocally found the more such control, the more product consistency and uniformity.

Product attribute number two: strength and elasticity. Polymers from single site catalysts tend to have high strength because they are linear or not branched. In making film, for example, this allows you to downgauge — make thinner films — that are strong enough to do the job and can readily incorporate comonomers needed to impart specific properties. This adds to the ability to make products that combine the best attributes of plastics and rubbers in single polymers.

Another attribute is the clarity of films made via single site catalysts. We're finding greater clarity than we get with conventional catalysts. Since improving clarity wasn't something we set out to achieve, we consider this a bit of serendipity. We're confident single site catalysts will produce more beneficial surprises in the future.

In describing other links in our technology chain, I mentioned the benefits of narrow MWD, narrow CD and high catalyst activities. These important attributes mean less likelihood of encountering problems associated with the taste and/or smell of components in polymer-based packaging that might be caused by leaching. We're finding superior organoleptic properties in products produced from polymers via single site catalysis.

The final product attribute I'd like to mention is thermal stability. When you melt polymers with broad molecular weight distribution, the low molecular weight components tend to be oily and smoky... the way motor oil smokes when a drop or tow touches the hot block of a car's engine. With the controlled molecular weight distribution, we can eliminate the low molecular weight components so this isn't likely to happen. Thus, you do away with the "smoking" other polymers can cause. That lack of low molecular weight components also contributes to better organoleptic properties as I just mentioned.

TECHNOLOGY DRIVES INDUSTRY TRENDS

- Licensing
- Alliances
- Profitability
- Technology synergy

In summary, it's apparent the development of single site catalysis is driving changes in polyolefin technology and this will cause significant industry changes.

There will be a delineation between licensors and licensees, and we'll see more and more teaming up among the players. Future growth and profitability of the polyolefin industry will be impacted and we'll see single site catalyst technology reaching beyond polyolefin polymers. And, interwoven in all this is tremendous synergy in technological developments sure to benefit not only the industry but our customers as well.

I close this preview of coming attractions with this suggestion: Reserve a day early in the second decade of the next century to attend a conference similar to this one. You may well hear about the latest once-in-every-20-year development in polyolefin technology. And I hope it will be my company telling you "how it went" with single site catalysis... and where our industry is heading next.

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Ultralow-density polyethylene

By L.D. Cady

Ultralow-density polyethylene and very low-density polyethylene are essentially synonymous terms referring to the category of linear polyethylenes with densities below 0.915 g./cc. Current technology provides a lower density limit between 0.880 and 0.890 g./cc.

The importance of these polymers stems from their unique combination of strength, sealability, flexibility, and optical properties. ULDPEs offer puncture, impact, tear, and optical properties superior to conventional linear low density polyethylenes (LLDPE), with sealing and flexibility properties comparable to 7 to 18% vinyl acetate copolymers (EVA).

Chemistry and properties

ULDPEs are manufactured by copolymerizing ethylene and selected alpha-olefins using a transition metal catalyst system. The polymers may employ a single alpha-olefin comonomer or a combination of comonomers.

The most common and commercially important comonomers are octene-1, hexene-1, butene-1, and 4-methylpentene-1. Comonomer content determines the density of the polymer, with increasing content resulting in lower density.

Additionally, it is well understood that employment of longer comonomers, i.e., octene-1 vs. butene-1, results in improvements in tear strength, puncture resistance, impact strength, and toughness of the polymer. This relationship is evident for conventional density LLDPEs, and enhanced for ULDPEs.

The lower density of ULDPE compared to other linear polyethylenes results in a softer, lower-modulus polymer. In turn, the lower modulus provides for better energy-absorbing characteristics of the polymer, which can yield superior impact and puncture resistance compared to LLDPEs. Additionally, ULDPEs are comparable in modulus to 7 to 18% vinyl acetate copolymers, but their linear nature results in significant advantages in impact, puncture, and tear resistance.

The softening point of linear polyethylene decreases as density decreases; therefore, the sealing range and minimum sealing temperature of ULDPE is significantly improved over LLDPE. ULDPE also offers excellent hot tack strength and sealability through contamination, both important attributes in film packaging applications.

The nonpolar nature of ULDPE offers better chemical resistance and thermal stability than polar copolymers; however, adhesion to polar substrates is generally inferior to that attainable by EVA. The adhesion of ULDPE to other polyethylenes, acid copolymers, and polypropylene is generally excellent.

The permeability of linear polyethylene is a function of the crystallinity of the polymer. Consequently, the lower crystallinity of ULDPE results in increased permeability to gases compared to higher density linear polymers, but similar to LDPE and EVA copolymers.

Processing

The rheological properties of ULDPE are similar to other linear polyethylenes, and dictated by the molecular weight and molecular weight distribution (MWD). A variety of grades ranging from fractional melt index to greater than 25 melt index, with narrow or broad MWDs, are available within this class of polymers.

ULDPEs can typically be extruded on existing polyethylene equipment, particularly equipment designed to process LLDPE. Modifications to high-shear, high-pressure equipment may be necessary to provide optimum processing of these polymers.

ULDPEs have very high thermal stability compared to polar copolymers. This results in the ability to run melt temperatures above 500°F., with the preferred range being 430 to 480°F.

In film manufacture, take-up and conversion of ULDPE is similar to EVA based on the modulus similarities. The toughness of ULDPE may also require improved slitting technology.

Applications

Most applications requiring superior toughness, puncture, tear, flexibility, and sealability are likely candidates for ULDPE. The unique combination of properties these polymers provide makes them suitable for film extrusion (especially coextrusion and lamination), blow molding, injection molding, sheet extrusion, filled systems, and profile and tubing applications. Additionally, several grades of these polymers conform to FDA regulations for direct food contact.

Specific film end-use applications include: meat, cheese, and poultry packaging, stretch film, bag-in-box, shrink film, heavy duty film, frozen food packaging, disposable gloves, medical packaging and film, and heat seal layers. Coextrusion and blending with HDPE and PP to improve tear and impact strength of those structures is also a significant application for these polymers.

Commercial information

ULDPE prices generally increase with decreasing density, reflecting increased comonomer costs. As of Jan. 1, 1988, film extrusion grades from 0.912 to 0.905 densities ranged in price from 60 to 65¢/lb., bulk railcar, respectively. □

L.D. Cady is Project Leader, Dow Chemical Co., Hwy. 288, Building B-1607, Freeport, TX 77541.

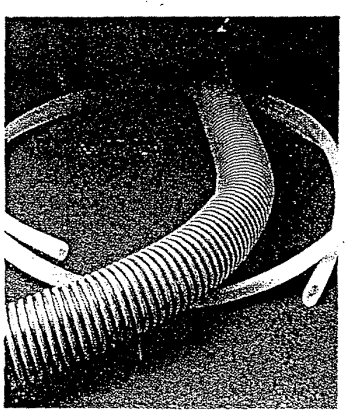

Fig. 1: ULDPE provides extruded tubing with flexibility over a range of temperatures, puncture resistance, and effective pigmentation. [Photo, Dow Chemical Co.]

High-density polyethylene

By Daniel D. Bussone

High-density polyethylene is a highly crystalline, lightweight thermoplastic. Outstanding characteristics are chemical resistance, toughness (even at low temperatures), dielectric properties, water vapor impermeability and relatively high softening temperature.

Manufacturing methods

There are three basic manufacturing techniques for the production of HDPE: particle form slurry reactors, tandem reactors, and gas phase reactors.

Particle form slurry methods, which are the most widely used processes, produce HDPE resins for all processing methods.

In particle form slurry processes, a supported transition metal catalyst slurried in a diluent is circulated in a closed-loop reactor at a low pressure and low temperature. Ethylene, which is dissolved in the diluent, reacts with the catalyst to form solid HDPE particles. The particles are dried, combined with additives, and extruded as pellets.

Tandem reactor methods have two particle form slurry reactors that are run in series or parallel, which permits short chain side branches to be regularly introduced into the linear molecular chains. This method yields lower-crystallinity resins having improved environmental stress cracking resistance, good water vapor permeability, impact strength, and enhanced processability.

Tandem reactors are used mainly to

Daniel D. Bussone is High Density Polyethylene Business Manager for Quantum Chemical Corp., USI Div., P.O. Box 429550, Cincinnati, OH 45249.

U.S. PTO
03/17/97

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RECEIVED
APR 4 1997
GROUP 1500

Applicant(s): Pak-Wing S. Chum, et al.

Serial No.: 08/544,497 Group Art Unit: 1505

Filed: October 19, 1995 Examiner: D. WU

For: FABRICATED ARTICLES MADE FROM ETHYLENE POLYMER BLENDS

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE AS FIRST CLASS MAIL WITH SUFFICIENT POSTAGE IN AN ENVELOPE ADDRESSED TO: COMMISSIONER OF PATENTS AND TRADEMARKS, WASHINGTON, D.C. 20231, On

March 12, 1997

DATE OF DEPOSIT

MaryAnn Navarrette

PRINT OR TYPE NAME OF PERSON SIGNING CERTIFICATE

MaryAnn Navarrette

SIGNATURE OF PERSON SIGNING CERTIFICATE

3-12-97

DATE OF SIGNATURE

Hon. Commissioner of Patents & Trademarks
Washington, D.C. 20231

Attn: Official Draftsman

Sir:

TRANSMITTAL OF FORMAL DRAWING(S)

Responsive to the Office Action (Paper No. 8) dated December 5, 1995, enclosed find the drawing(s) required for the understanding of the subject matter of the invention in compliance with 35 USC §113.

Respectfully submitted,

By

Osborne K. McKinney
Registration No. 40,084
Address: Bldg. B-1211
Freeport, Texas 77541
Phone: 409-238-7889

OKM/man

40121AU

APPROVED BY DRAFTSMAN
G FIG.
CLASS SUBCLASS
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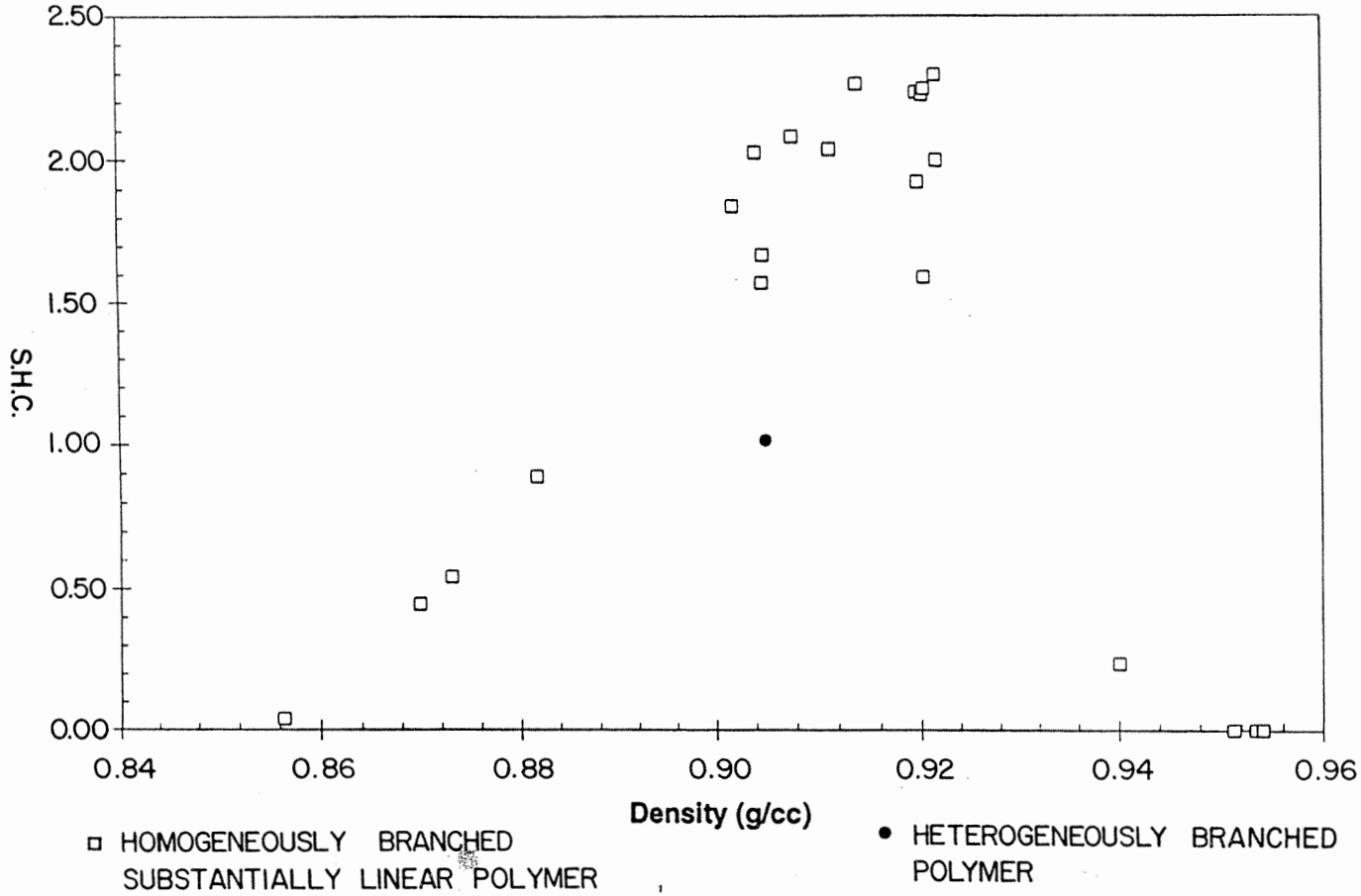
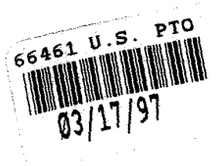
FIG. 1





40121AU
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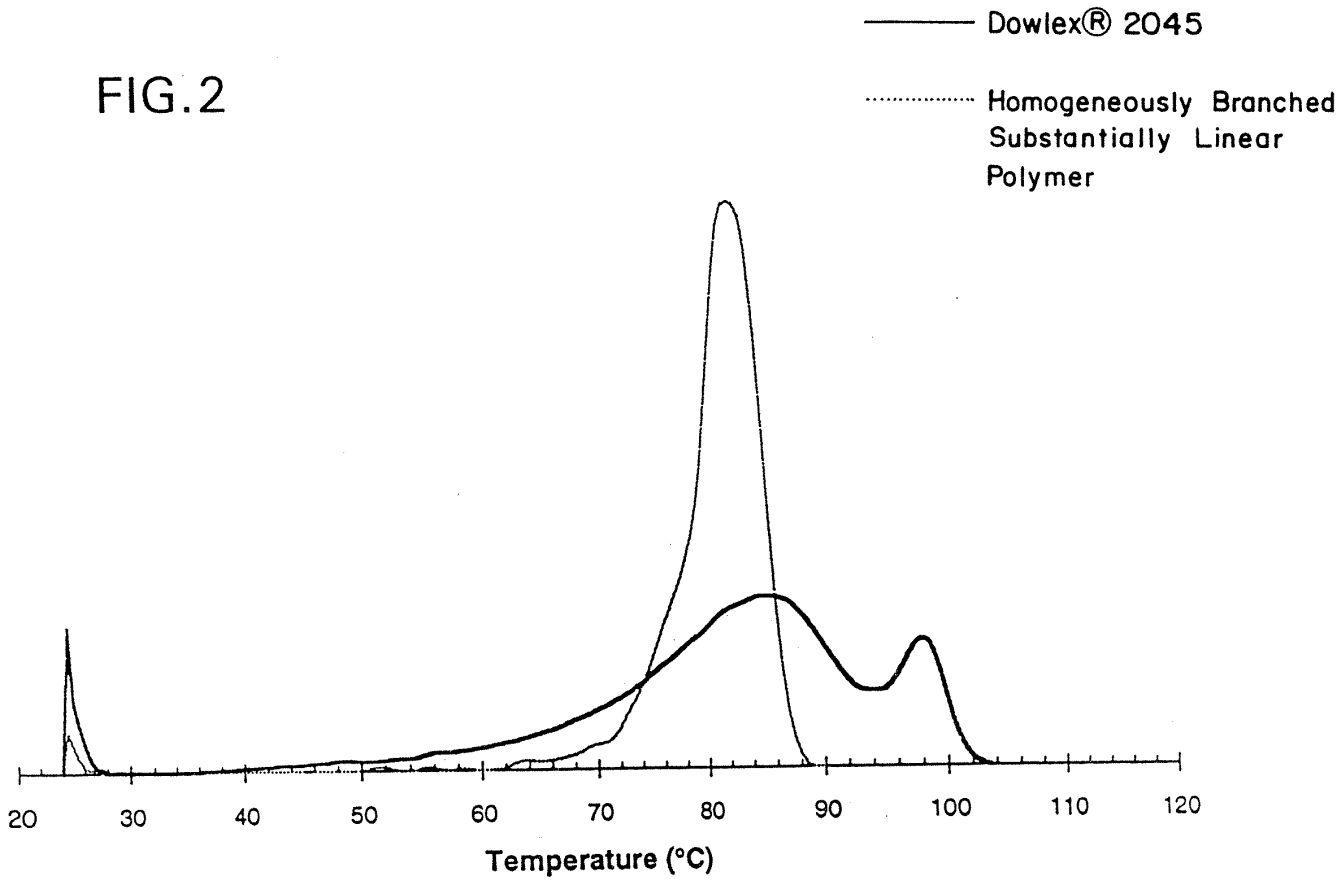
Osborne K. McKinney
(409) 238-7889
Sheet 1 of 1

APPROVED BY DRAFTSMAN
FIG
CLASS SUBCLASS

FIG. 2



66461 U.S. PTO
03/17/97

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08/544,497
1505
Osborne K. McKinney
(409) 238-7889
Sheet 1 of 2

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APR 4 1997
GROUP 1500

Form PTO 948 (Rev. 10-94) U.S. DEPARTMENT OF COMMERCE - Patent and Trademark Office Application No. 544497

NOTICE OF DRAFTSPERSON'S PATENT DRAWING REVIEW

PTO Draftpersons review all originally filed drawings regardless of whether they are designated as formal or informal. Additionally, patent Examiners will review the drawings for compliance with the regulations. Direct telephone inquiries concerning this review to the Drawing Review Branch, 703-305-8404.

The drawings filed (insert date) 10/18/95, are
A. ___ not objected to by the Draftsperson under 37 CFR 1.84 or 1.152.
B. X objected to by the Draftsperson under 37 CFR 1.84 or 1.152 as indicated below. The Examiner will require submission of new, corrected drawings when necessary. Corrected drawings must be submitted according to the instructions on the back of this Notice.

1. DRAWINGS. 37 CFR 1.84(a): Acceptable categories of drawings: Black ink. Color.
   ___ Not black solid lines. Fig(s)___
   ___ Color drawings are not acceptable until petition is granted. Fig(s) ___
2. PHOTOGRAPHS. 37 CFR 1.84(b)
   ___ Photographs are not acceptable until petition is granted. Fig(s) ___
   ___ Photographs not properly mounted (must use brystol board or photographic double-weight paper). Fig(s)___
   ___ Poor quality (half-tone). Fig(s)___
3. GRAPHIC FORMS. 37 CFR 1.84 (d)
   ___ Chemical or mathematical formula not labeled as separate figure. Fig(s)___
   ___ Group of waveforms not presented as a single figure, using common vertical axis with time extending along horizontal axis. Fig(s)___
   ___ Individuals waveform not identified with a separate letter designation adjacent to the vertical axis. Fig(s)___
4. TYPE OF PAPER. 37 CFR 1.84(c)
   ___ Paper not flexible, strong, white, smooth, nonshiny, and durable. Sheet(s)___
   X Erasures, alterations, overwritings, interlineations, cracks, creases, and folds copy machine marks not accepted. Fig(s) 1
   ___ Mylar, velum paper is not acceptable (too thin). Fig(s) ___
5. SIZE OF PAPER. 37 CFR 1.84(f): Acceptable sizes:
   21.6 cm. by 35.6 cm. (8 1/2 by 14 inches)
   21.6 cm. by 33.1 cm. (8 1/2 by 13 inches)
   21.6 cm. by 27.9 cm. (8 1/2 by 11 inches)
   21.0 cm. by 29.7 cm. (DIN size A4)
   ___ All drawing sheets not the same size. Sheet(s)___
   ___ Drawing sheet not an acceptable size. Sheet(s) ___
6. MARGINS. 37 CFR 1.84(g): Acceptable margins:

Paper size

21.6 cm. X 35.6 cm. (8 1/2 X 14 inches)	21.6 cm X 33.1 cm. (8 1/2 X 13 inches)	21.6 cm. X 27.9 cm. (8 1/2 X 11 inches)	21.0 cm. X 29.7 cm. (DIN Size A4)
T 5.1 cm. (2")	2.5 cm. (1")	2.5 cm. (1")	2.5cm.
L .64 cm. (1/4")	.64 cm. (1/4")	.64 cm. (1/4")	2.5 cm.
R .64 cm. (1/4")	.64 cm. (1/4")	.64 cm. (1/4")	1.5 cm.
B .64 cm. (1/4")	.64 cm. (1/4")	.64 cm. (1/4")	1.0 cm.

Margins do not conform to chart above. Sheet(s) 2
___Top (T) X Left (L) ___Right (R) ___Bottom (B)

7. VIEWS. 37 CFR 1.84(h)
   REMINDER: Specification may require revision to correspond to drawing changes.
   ___ All views not grouped together. Fig(s)___
   ___ Views connected by projection lines or lead lines. Fig(s)___
   Partial views. 37 CFR 1.84(h) 2
   ___ View and enlarged view not labled separatly or properly. Fig(s)___
   Sectional views. 37 CFR 1.84 (h) 3
   ___ Hatching not indicated for sectional portions of an object. Fig(s)___
   ___ Cross section not drawn same as view with parts in cross section with regularly spaced parallel oblique strokes. Fig(s)___
8. ARRANGEMENT OF VIEWS. 37 CFR 1.84(i)
   ___ Words do not appear on a horizontal, left-to-right fashion when page is either upright or turned so that the top becomes the right side, except for graphs. Fig(s)___
9. SCALE. 37 CFR 1.84(k)
   ___ Scale not large enough to show mechanism with crowding when drawing is reduced in size to two-thirds in reproduction. Fig(s)___
   ___ Indication such as "actual size" or scale 1/2" not permitted. Fig(s)___
10. CHARACTER OF LINES, NUMBERS, & LETTERS. 37 CFR 1.84(l)
   ___ Lines, numbers & letters not uniformly thick and well defined, clean, durable, and black (except for color drawings). Fig(s)___
11. SHADING. 37 CFR 1.84(m)
   ___ Solid black shading areas not permitted. Fig(s)___
   ___ Shade lines, pale, rough and blurred. Fig(s)___
12. NUMBERS, LETTERS, & REFERENCE CHARACTERS. 37 CFR 1.84(p)
   ___ Numbers and reference characters not plain and legible. 37 CFR 1.84(p)(l) Fig(s)___
   ___ Numbers and reference characters not oriented in same direction as the view. 37 CFR 1.84(p)(l) Fig(s)___
   ___ English alphabet not used. 37 CFR 1.84(p)(2) Fig(s)___
   ___ Numbers, letters, and reference characters do not measure at least .32 cm. (1/8 inch) in height. 37 CFR(p)(3) Fig(s)___
13. LEAD LINES. 37 CFR 1.84(q)
   ___ Lead lines cross each other. Fig(s)___
   ___ Lead lines missing. Fig(s)___
14. NUMBERING OF SHEETS OF DRAWINGS. 37 CFR 1.84(t)
   ___ Sheets not numbered consecutively, and in Arabic numerals, beginning with number 1. Sheet(s)___
15. NUMBER OF VIEWS. 37 CFR 1.84(u)
   ___ Views not numbered consecutively, and in Arabic numerals, beginning with number 1. Fig(s)___
   ___ View numbers not preceded by the abbreviation Fig. Fig(s)___
16. CORRECTIONS. 37 CFR 1.84(w)
   ___ Corrections not made from prior PTO-948. Fig(s)___
17. DESIGN DRAWING. 37 CFR 1.152
   ___ Surface shading shown not appropriate. Fig(s)___
   ___ Solid black shading not used for color contrast. Fig(s)___

COMMENTS:

REVIEWER [illegible] DATE 12/5/95

PATENT APPLICATION FEE DETERMINATION RECORD

Effective October 1, 1995

Application or Docket Number: 08 544497

CLAIMS AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA
BASIC FEE		
TOTAL CLAIMS	30 minus 20 =	* 10
INDEPENDENT CLAIMS	4 minus 3 =	* 1
MULTIPLE DEPENDENT CLAIM PRESENT		

* If the difference in column 1 is less than zero, enter "0" in column 2

SMALL ENTITY RATE	SMALL ENTITY FEE	OR	OTHER THAN SMALL ENTITY RATE	OTHER THAN SMALL ENTITY FEE
	375.00	OR		750.00
x$11=		OR	x$22=	220
x39=		OR	x78=	78
+125=		OR	+250=	
TOTAL		OR	TOTAL	1048

CLAIMS AS AMENDED - PART II

AMENDMENT A

	(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA
Total	* 15	Minus	** 30	= —
Independent	* 2	Minus	*** 4	= —
FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM				

SMALL ENTITY RATE	SMALL ENTITY ADDITIONAL FEE	OR	OTHER THAN SMALL ENTITY RATE	OTHER THAN SMALL ENTITY ADDITIONAL FEE
x$11=		OR	x$22=	
x39=		OR	x78=	
+125=		OR	+250=	
TOTAL ADDIT. FEE		OR	TOTAL ADDIT. FEE	

AMENDMENT B

	(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA
Total	* 18	Minus	** 30	= —
Independent	* 6	Minus	*** 4	= 2
FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM				

SMALL ENTITY RATE	SMALL ENTITY ADDITIONAL FEE	OR	OTHER THAN SMALL ENTITY RATE	OTHER THAN SMALL ENTITY ADDITIONAL FEE
x$11=		OR	x$22=	
x39=		OR	x78=	156
+125=		OR	+250=	
TOTAL ADDIT. FEE		OR	TOTAL ADDIT. FEE	

AMENDMENT C

	(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA
Total	*	Minus	**	=
Independent	*	Minus	***	=
FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM				

SMALL ENTITY RATE	SMALL ENTITY ADDITIONAL FEE	OR	OTHER THAN SMALL ENTITY RATE	OTHER THAN SMALL ENTITY ADDITIONAL FEE
x$11=		OR	x$22=	
x39=		OR	x78=	
+125=		OR	+250=	
TOTAL ADDIT. FEE		OR	TOTAL ADDIT. FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20."
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3."
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.

FORM PTO-875
(Rev. 10/95)

Patent and Trademark Office, U.S. DEPARTMENT OF COMMERCE

Case 1:05-cv-00737-JJF Document 160-9 Filed 09/03/2008 Page 47 of 52

EXHIBIT PAGE 001196

Form PTO 1130
(REV 2/94)

U.S. DEPARTMENT OF COMMERCE
Patent and Trademark Office

PACE DATA ENTRY CODING SHEET

1ST EXAMINER: Jissetts — DATE: 11-28-95

2ND EXAMINER: — DATE:

APPLICATION NUMBER	TYPE APPL	FILING DATE (MONTH DAY YEAR)	SPECIAL HANDLING	GROUP ART UNIT	CLASS	SHEETS OF DRAWING
08/544497	1	10 18 95	2	1505	526	2

TOTAL CLAIMS	INDEPENDENT CLAIMS	SMALL ENTITY?	FILING FEE	FOREIGN LICENSE	ATTORNEY DOCKET NUMBER
30	4	0	1084	Y	C-40.1210-AU

CONTINUITY DATA

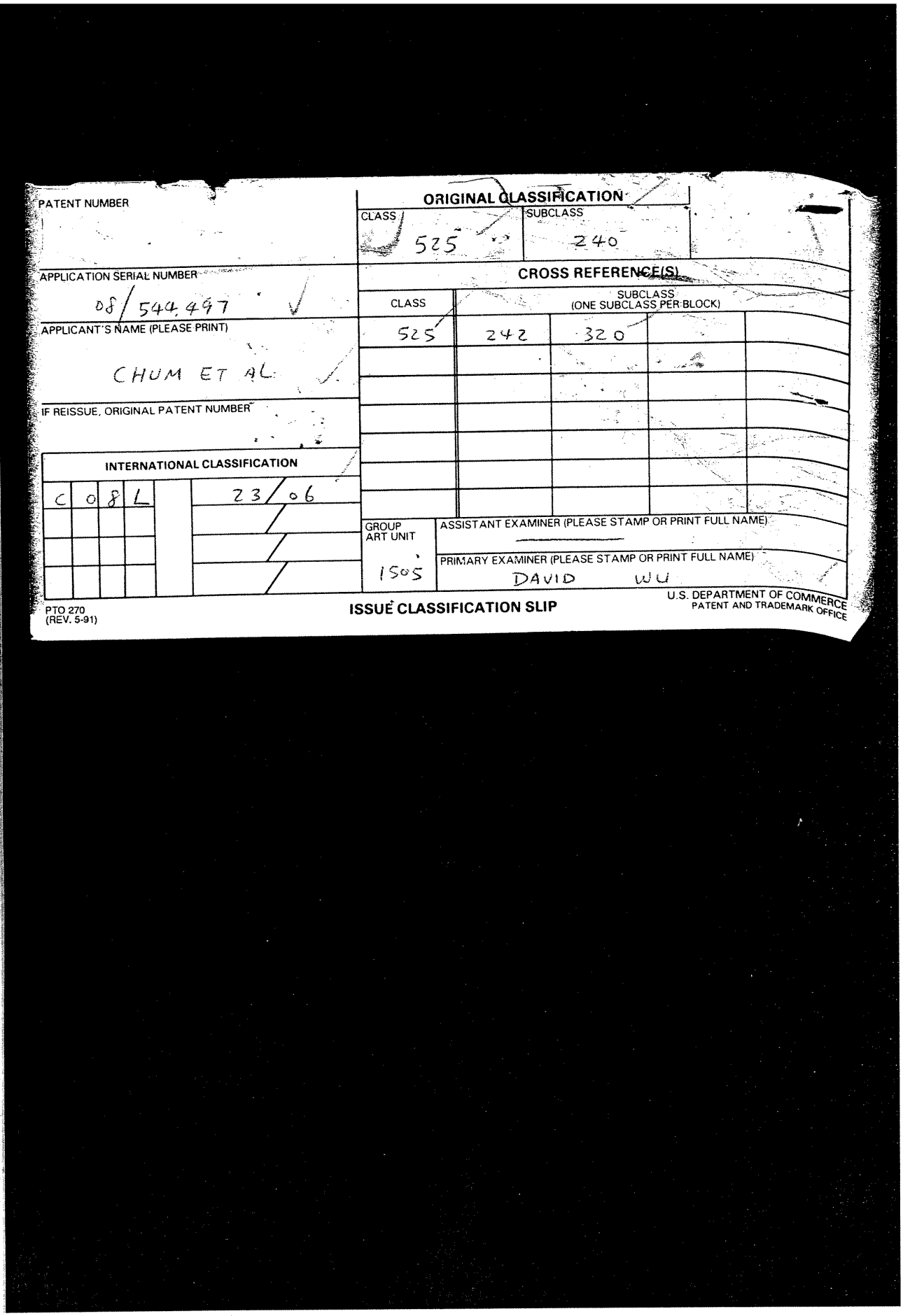
CONT CODE	STATUS CODE	PARENT APPLICATION SERIAL NUMBER	PCT APPLICATION SERIAL NUMBER	PARENT PATENT NUMBER	PARENT FILING DATE (MONTH DAY YEAR)
02	2	08/378,998	PCT/		01/27/95
13	2	07/776,130	PCT/		10/15/91
			PCT/		
			PCT/		
			PCT/		

PCT/FOREIGN APPLICATION DATA

FOREIGN PRIORITY CLAIMED	COUNTRY CODE	PCT/FOREIGN APPLICATION SERIAL NUMBER	FOREIGN FILING DATE (MONTH DAY YEAR)

☆U.S.G.P.O.: 1994 - 384-021

CITY
GIVEN NAME
FAMILY NAME
ATTORNEY CODE
CITY
GIVEN NAME
FAMILY NAME
ATTORNEY CODE
APPLICANT/INVENTOR DATA
CORRESPONDENCE AND ADDRESS
ATTORNEY DOCKET NUMBER
TITLE OF INVENTION
MORE
NAME SUFFIX
STATE/CTRY CODE
NAME SUFFIX
STATE/CTRY CODE



PATENT NUMBER

APPLICATION SERIAL NUMBER
08/544,497

APPLICANT'S NAME (PLEASE PRINT)
CHUM ET AL

IF REISSUE, ORIGINAL PATENT NUMBER

INTERNATIONAL CLASSIFICATION
C 08 L 23/06

ORIGINAL CLASSIFICATION

CLASS	SUBCLASS
525	240

CROSS REFERENCE(S)

CLASS	SUBCLASS (ONE SUBCLASS PER BLOCK)			
525	242	320		

GROUP ART UNIT
1505

ASSISTANT EXAMINER (PLEASE STAMP OR PRINT FULL NAME)

PRIMARY EXAMINER (PLEASE STAMP OR PRINT FULL NAME)
DAVID WU

PTO 270
(REV. 5-91)

ISSUE CLASSIFICATION SLIP

U.S. DEPARTMENT OF COMMERCE
PATENT AND TRADEMARK OFFICE

Staple Issue Slip Here

POSITION		ID NO.	DATE
CLASSIFIER		5	11-28-95
EXAMINER		353	11-28-95
TYPIST		284	11/28
VERIFIER		441	11/28
CORPS CORR.			
SPEC. HAND			
FILE MAINT.			
DRAFTING			

INDEX OF CLAIMS

Claim Final	Claim Original	Date 96 8	Date 96 12 13
1	(1)	✓	=
	2		
2	3	✓	=
5	4	|	|
6	5	|	|
7	6	|	|
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SYMBOLS

✓ ..... Rejected
= ..... Allowed
- (Through numberal) Canceled
+ ..... Restricted
N ..... Non-elected
I ..... Interference
A ..... Appeal
O ..... Objected

Claim Final	Claim Original	Date
	51	
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SEARCHED

Class	Sub.	Date	Exmr.
525	240	8-2-96	WH
	242		
	320		
	ATD	12-13-96	WH

SEARCH NOTES

	Date	Exmr.
NDH		WH

INTERFERENCE SEARCHED

Class	Sub.	Date	Exmr.
525	240		
	242		
	320	12-13-96	WH

(RIGHT OUTSIDE)

4497

PATENT APPLICATION

08544497

PROVED FOR LICENSE

TIALS

No. 28 9 5 31

Date Entered or Counted

CONTENTS

Date Received or Mailed

RECEIVED DEC 12 1995

1.	Application _____ papers.	
2.	Pre Amdt A	10-18-95
3.	Pre Amdt B	Feb 6, 1996
4.	Pre Amdt C	Feb 6 1996
5.	Prim Art	Mar 13, 1996
6.	Suppl Oath	Mar 13 1996
7.	Prim Art	Apr 10, 1996
8.	Rej 3 mos	AUG 12 1996 8/2
8½	Interview Summary	10/9/96
9.	Amdt D Yatch	10/9/96
10.	~~Suppl~~ Notice of Allowance	DEC 19 1996 12/12
11.	PTO Grant OCT 14 1997	
12.	Req. CofC	6/16/06
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