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PREVENTION OF GEL FORMATION IN RECYCLED LLDPE PROCESS

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ภาควิชา ปิโตรเคมีและวิทยาศาสตร์พอลิเมอร์ **ลายมือชื่อนิสิต**...
สาขาวิชา ปิโตรเคมีและวิทยาศาสตร์พอลิเมอร์ **ลายมือชื่ออาจารย์ที่ปรึกษา**................................
ปีการศึกษา 2544

4173411523 : MAJOR PETROCHEMISTRY AND POLYMER SCIENCE
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In this research, scrap of LLDPE film after processing from blown film process will be recycled by the addition of antioxidant at various concentrations. Then, the effects of its concentration and efficiency related to reduction of gel contents on film was studied. The compounded masterbatch of phenolic antioxidant, octadecyl 3,5-bis(1,1-dimethylethyl)-4-hydroxybenzene propanoate (Irganox 1076) and phosphite type, tris(2,4-di-tert-butylphenyl)phosphite (Irgafos 168) as it called polybatch AO-25 masterbatch was blended with scrap LLDPE at concentration of 3, 8, 15, 25, 40 phr. Furthermore, each formulation was used to prepare a film sheet using the blown film process and the gel counting, mechanical properties and yellowness index (YI) were studied. It was found that addition of polybatch AO-25 reduced the gel contents when compared with unfilled formula. It was observed that there was no longer decreasing of gels on film when polybatch AO–25 was at 15 to 40 phr. However, the value of mechanical properties and yellowness index became less and more when the contents of polybatch AO-25 was increasing. Consequently, addition of virgin grade LLDPE into these formulations could be a solution. From the testing results, using polybatch AO-25 8 phr blended with 25 % virgin grade LLDPE was the most effective to provide low gel contents, good mechanical properties and low yellowness index.

Department Petrochemistry and Polymer Science Student's signature…….……
Field of study Petrochemistry and Polymer Science Advisor's signature………….....
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CONTENTS

PAGE

CONTENTS (continued)

PAGE

CONTENTS (continued)

PAGE

LIST OF TABLES

LIST OF FIGURES

LIST OF SCHEMES

CHAPTER I

INTRODUCTION

1.1 Background

LLDPE (linear low density polyethylene), a new addition to polymer family is making both the users and the converters sit up and take a second look. The prospects look rather bright because the material has all the advantages of the family low density polyethylene together with the added benefits of low energy input which leads to manufacturing cost saving of over 20 percents. This material also offers a cost advantage over low density polyethylene (LDPE) in lesser material consumption as 30-50 percents downgauging is possible in the case of polymer film without compromising in any way the mechanical properties and in most of the cases even improving them in the process (Table 1.1) [1].

LLDPE is one of the fastest growing polymers at 15-20 percents growth per year of Western Europe in the context of volume and usage. The biggest areas of growing use are flexible packaging, heavy-duty bags and ice bags. At present, 65-70 % of LLDPE use is in film application. Blown film is the most important method for producing LLDPE film. It is estimated that some 90 % of LLDPE films are produced on blown film lines[2]. Normally blown film process has many scraps because it is from set up or change of sizing. LLDPE scrap from process has been recycled by reprocessed to film reclaiming. Film reclaiming has many problems, for example :

1) Reduction in physical properties
2) Presence of many odors
3) Increasing of yellowness
4) Presence of gel

Table 1.1 Comparison of LDPE and LLDPE blown film physical characteristics[2]

Property	ASTM Test		Type of Polyethylene (PE)				
			HP-LDPE		LLDPE		
Melt index	-		2.5	0.2	1.0	1.0	1.0
Density, g/cm^3	-		0.921	0.923	0.918	0.918	0.920
Comonomer	-		none	none	butene	hexene	octene
Dart drop, N/mm	D 1709		29.0	71.4	38.6	77.2	96.5
Puncture energy, kJ/m	-		26.7	22.2	71.2	75.6	-
Elmendorf tear, N/mm	D 1922	MD	61.8	34.7	54.0	131.2	142.8
		TD	42.5	38.6	131.2	225.8	308.8
Tensile strength, Mpa	D 882	MD	20.0	19.3	34.5	35.9	44.8
		TD	18.6	20.7	26.2	32.4	35.2
Tensile impact strength, Mpa	-	MD	36.4	41.4	99.3	159.7	-
		TD	53.8	86.9	74.5	145.6	-

A gel is the result of a disruption in the polymer flow as the film is fabricated. The key to understanding how to correct a gel problem is to first identify the type of gel : [3]

1. Contamination gels: Some of the common sources of contamination gels are cellulose, cotton, dirt, dust, and insect etc.
2. Polymer contamination gels : Generally occur from extruding a different polymer from that which is intended.The mixing of polymers with widely different melt index or molecular weight can result in gels due to incompatibilities in rheological flow characteristic.
3. Unmelt /unmixed gels : These types of gels are due to incomplete melting, mixing and dispersion in the extruder. Typical sources are poorly dispersed additives.

4. Polymer degradation gels: There are two types according to sources of formation.
    - 4.1 Crosslinking gels: They are formed from free radicals for crosslinking to be higher molecular weight product.
    - 4.2 Oxidized gels: They are undesired oxidation reactions occur in the presence of oxygen.

1.2 Objective

The focus of this research is on the reduction of gel formation in recycled linear low density polyethylene (LLDPE) film by using various amount of antioxidant.

1.3 Scope of the investigation

The scope of this research work involves the blending of scrap LLDPE film with various amount of virgin LLDPE and antioxidant masterbatch using single screw extruder and then blown film machine to form recycled LLDPE film which reduced amount of gel content. The pellets from single screw extruder were also measured for the melt flow index (MFI) in order to find the effect on blown film process .

Investigation of the number of gel present in recycled LLDPE film was performed, according to ASTM D 3351-74. In addition, tensile strength, according to ASTM D 882-91, tear resistance, according to ASTM D 1938-92, and yellowness index, according to ASTM D 1925-70, were also observed for indication of recycled LLDPE film properties.

CHAPTER II

THEORY

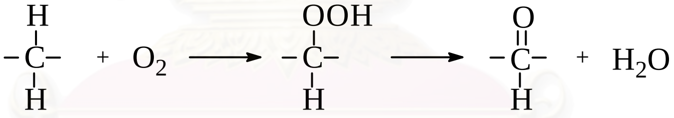
2.1 Theoretical background

2.1.1 Degradation of plastic

During the manufacture processing of polymer material as well as its fabrications into an article it will be subjected to a variety of degradative processes which will itself be dependent upon the nature of the polymer and its end-use. Degradation usually manifests itself as:

1.) Discoloration

Yellowing development in degradation is attributed to transformations of hydroperoxide resulting from heating to be ketone group which cause oxidation of polymer [4].



$$-\overset{H}{\underset{H}{C}}- + O_2 \longrightarrow -\overset{OOH}{\underset{H}{C}}- \longrightarrow -\overset{O}{\underset{H}{C}}- + H_2O$$

2.) *Loss of volatile components (smoking)*

A.Bravo and J.H.hotchkiss separated and identified the products of LDPE decomposition from the thermo-oxidative degradation of LDPE at a low degree of volatiliation at 264-289 ^{o}C. Altogether were compounds and in some parts were aldehyde and ketone can present the smelling [5].

3.) Loss of machanical properties

S.J. Hebert and C. Tryogorakis studied the properties of blown film made from post consumer recycled (PCR) material. The tear strength and dart drop impact strength can be seen that there is a significant decrease at increasing levels of PCR [6].

4.) Gel

Gel is a crosslinked by more molecular chains come together.

2.1.2 Sources of gel

Definition of a gel is the result of a disruption in the polymer flow as the film is fabricated[1]. Gel can be happened by many causes. There are three main categories for the gel formation[7].

1.) Polymer

The gel formation from polymer resulted from the present of high molecular weight polymer, structural changing by degradation and poor additives dispersion.

2.) Equipment and process

Equipment design for used in the process can create the gel formation and difficult to control of gels.

3.) Processing condition

In this case, the operating procedures and conditions for used in the process cause the changes in the polymer structure, for example temperature, start-up and purging procedures [5].

2.1.3 Identification of gel

Gel can be happened in conventional polymer processing and it has many types. The key to understanding how to correct a gel problem is to first identify the type of gel as shown below[8]:

1.) *Contamination gel*

It can come from dust, dirt and insects, etc.

2.) *Polymer contamination*

Some polymer could be a high molecular weight and can present a different melting temperature in polymer blend.

3.) *Unmixed or unmelt polymer*

Unmelt gels are believed to form from small domains of polymer at a lower temperature and it can come from many sources, for example,

- Poor dispersion of additive.
- Broad range of polymer melting.
- Non-uniform melting of solids.
- Break-up of solids bed.

The degradation problem of unmelt gel mostly come from equipment design or operating procedures.

4.) *Polymer degradation*

Polymer can change its fabrication during the manufacture or processing and it resulted in gel formation from this degradation. This gel type is divided into two categories as shown below:

4.1) Crosslinked gel

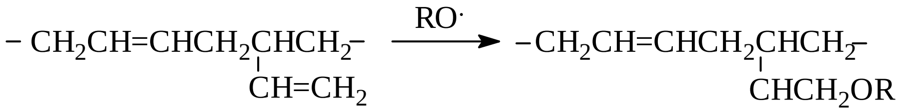
Gel forms free radicals for crosslinking from unsaturation during processing[9] :

$$2\sim[CH_2-CH_2]\sim \xrightarrow{\triangle} \begin{array}{c}\sim[CH-CH_2]\sim\\ |\\ \sim[CH-CH_2]\sim\end{array}$$

Polymer will present more or less gel depended on three factors in processing which are residence time, temperature and shearing. Crosslinked gels is undesirable reactions which form from large molecules when polymer is exposed to at high temperatures over long period of time.

4.2) Oxidized gel

This oxidation reaction occurs very fast compared to crosslinked rate and undesirable oxidation reactions occur in the presence of oxygen which only small amounts of oxygen can propagate the reaction. The oxidized gel reaction in polybutadiene rubber can be shown below[9]:



$$-CH_2CH{=}CH\underset{\displaystyle CH{=}CH_2}{CH_2CH}CH_2- \xrightarrow{RO^{\cdot}} -CH_2CH{=}CH\underset{\displaystyle CHCH_2OR}{CH_2CH}CH_2-$$

This reaction caused by alkoxy radical which was generated from oxidation cycle (Scheme 2.1) [7].

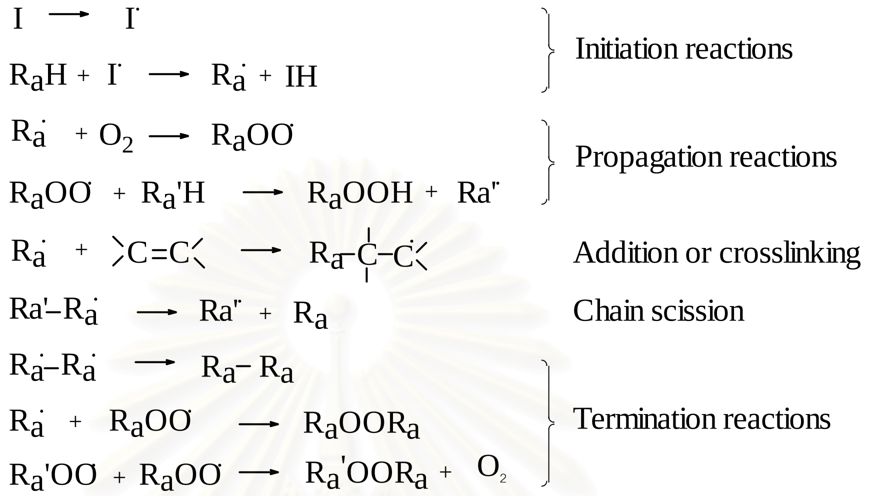
2.1.4 Oxidation reaction

Although all polymers degrade at high temperatures in the absence of air, degradation is almost always faster in the presence of oxygen. Oxidation of hydrocarbons is normally auto-accelerating, i.e. the rate is slow or even regligible at first but gradually acceleration, often to a constant value. The addition of an initiator normally removes the slow auto accelerating induction time and antioxidants while stabilizers extend it.

The process of oxidation is rather complex, but the following generalized Scheme 2.1 demonstrates the most important features of autoxidation processes.

Any oxidation of polymer requires an initiation process, which generates free radical from the substrate. These free radicals are able to react with atmospheric oxygen, a particularly facile reaction resulting from the diradical nature of the oxygen molecule, to give a peroxy radical. This peroxy radical is very reactive and will rapidly abstract a hydrogen atom from the surrounding medium or substrate to yield a new free radical and a hydroperoxide molecule. Since a new free radical is generated at each step, more oxygen is incorporated into the system as this chain reaction proceeds, and the reaction is terminated only when two free radicals combine to form a nonradical product [4].

During the course of the chain reaction chain scission can occur, usually with the introduction of an oxygen function into the molecule which often acts as a photosensitizer, absorbing light or ultraviolet radiation and generating further free radicals to initiate further oxidation, introduction discoloration and, most important breaking down the substrate molecules. In unsaturated systems, the free radical can add to centers of unsaturation, generating a new free radical and joining two substrate molecules together. These are two of the destructive elements of oxidation, causing gross changes in the chemical and physical properties of the substrate.



I or IH is a species which forms the free radicals I and R_aH is an polymer.

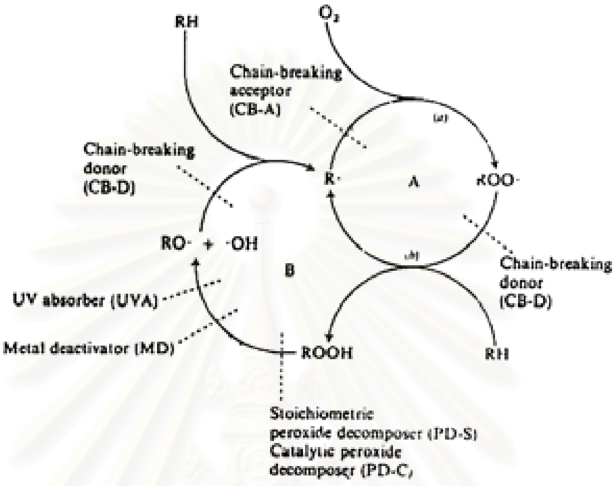
Scheme 2.1 The oxidation reaction process

The hydroperoxide molecules generated in the propagation reaction are thermally unstable, and generally undergo heterolytic fission to yield two free radicals. These are able to initiate further chain reaction, and so the whole process may be said to be autocatalytic. In order to suppress oxidation, the addition of an antioxidant is required, and this may function in several ways, either interfering with the chain reaction mechanism or by suppressing initiation reactions [4].

2.1.5 Mechanisms of antioxidant

Antioxidants are inhibitors for autoxidation. In the present the term will be used to include heat stabilisers, melt stabilisers, light (UV) stabilisers, antifatigue agents and antiozonants. All these agents interfere with the free radical reactions that lead to the incorporation of oxygen into macromolecules.

The autoxidation chain mechanism is summarized in Scheme 2.2 [9]. It involves two interacting cyclical processes. The first cycle is the alkyl/ alkylperoxyl chain reaction and the second involves the homolysis of hydroperoxides which feeds the chain reaction with new radicals. In the stationary state, the rate of formation of hydroperoxides by the A cycle is equal to their rate of decomposition by the B cycle and the rate of oxidation is constant .

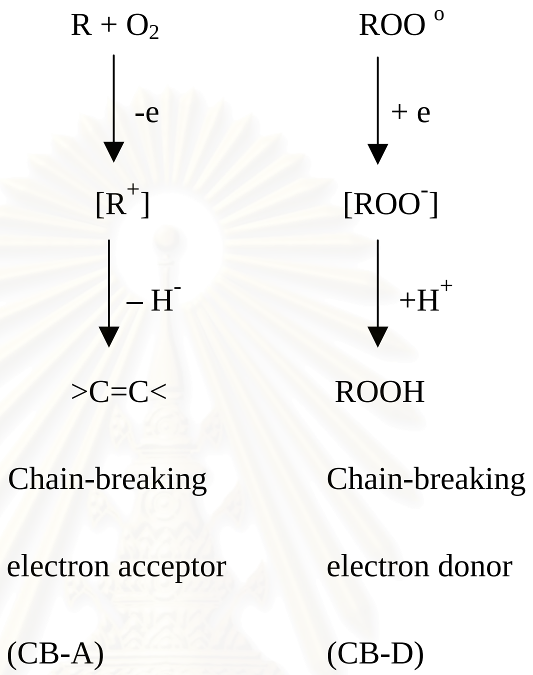


Scheme 2.2 Mechanisms of antioxidant action

The purpose of adding an inhibitor is to prevent this stage being reached by inhibiting or retarding the formation of hydroperoxides for as long as possible. Scheme also indicates the points at which anti-oxidants can in principle operate.

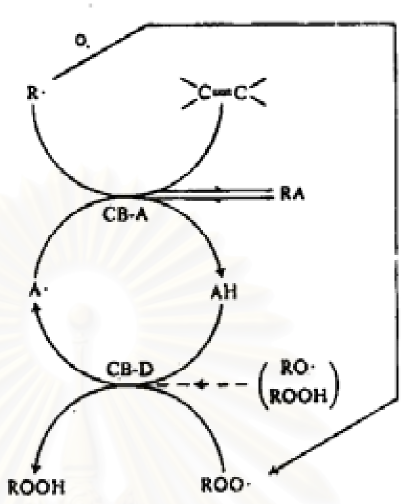
2.1.5.1 Chain-breaking antioxidants

The primary cycle can be interrupted at two points. In the presence of an oxidizing agent the alkyl radical can be removed to give a carbonium ion and subsequent inert reaction products such as olefins by elimination of a proton. The alkylperoxyl radical can be reduced to give a hydroperoxide. Antioxidants acting by these two mechanisms are electron acceptors (oxidizing agents) and electron donors (reducing agents) respectively, and will be designated as CB-A and CB-D antioxidants for convenience in the Scheme 2.3 [10] as followed:



Scheme 2.3 Kinetic chain-breaking mechanisms.

If both alkyl and alkylperoxyl radicals are present in an autoxidising system in comparable concentration at the same time, then both CB-A and CB-D mechanisms may operate simultaneously. Moreover, if the oxidised and reduced forms of an antioxidant form a reversible redox couple, then more than one kinetic chain may be broken per antioxidant molecule. Under these favourable conditions catalytic inhibition may occur and is generally represented in scheme. It can be seen that the catalytic antioxidant cycle in Scheme 2.4 is in competition with the catalytic oxidation cycle A of Scheme 2.2. It was seen earlier that in most autoxidantions the rate determining step in Scheme 2.2 is reaction (b). Consequently the CB-A radical, so that only the CB-D mechanism can terminate chain propagating radicals efficiently.



Scheme 2.4 General catalytic chain-breaking mechanism of antioxidant action [11]

2.1.5.2 Preventive antioxidants

Antioxidants acting on the B cycle are “preventive” since their primary function is to interfere with the generation of free radicals which feed back into the A cycle. The most important preventive mechanism, both theoretically and practically, is hydroperoxide decomposition (PD) in a process which does not involve the formation of free radicals. Proxidolytic antioxidants fall into two classes; stoichiometric peroxide decomposers (PD-S), such as phosphite esters which are reagents for the reduction of hydroperoxides to alcohol, and catalytic peroxide decomposers (PD-C). A variety of sulphur compounds that are of considerable importance as commercial anti-oxidants fall into the latter class and their mechanisms have been extensively studied in recent years.

Although complete destruction of hydroperoxides from the manufacture of the polymer artifact through its service life is the ideal to be aimed at, other agents that slow down their decomposition may act as retarders of peroxide initiated oxidation. Many transition mental ions are effective catalysts for the homolytic decomposition of hydroperoxides. They are therefore effective pro-oxidants and their activity depends on the availability of oxidized and reduced states of comparable stability. Complexing agents which have the ability to co-ordinate the vacant orbitals of transition metal ions to their maximum co-ordination number and thus inhibit the co-ordination of hydroperoxides to the metal ions are therefore effective metal deactivators (MD) and hence inhibitors for metal catalysed autoxidation.

Similarly in the case of UV-initiated oxidation, UV light absorbers (UVA), screens, filters, etc, effectively inhibit the photo-excitation of light absorbing species present in the polymer. Another way, in which preventive antioxidants may in principle act, is by quenching of photo-excited states of chromophores present in polymers (e.g. carbonyl compounds). In practice, many commercial stabilizers have been found to operate by a combination of these mechanisms.

2.1.5.3 Synergism and antagonism

Most stabilizers for polymers contain a combination of anti-oxidants acting by different and normally complementary mechanisms. It will be clear from the above discussion that an antioxidant which destroys hydroperoxides, thereby reducing the concentration of radicals in the a cycle will in consequence slow down the destruction of a chain-breaking antioxidant. By the same token, an effective chain-breaking antioxidant reduces the amount of hydroperoxide formed in an autoxidising system and hence protects a peroxide decomposer. This co-operative interaction, which is commonly called synergism, leads to an overall antioxidant effect which is greater than the sum of the individual effects and very often to an effectiveness much greater than can be achieved by either antioxidant alone even at much higher concentrations. The phenomenon is therefore of considerable practical and theoretical significance.

Occasionally the reverse phenomenon is observed; that is, two antioxidants interact to decrease the sum of their individual effects. This is described as antagonism.

2.1.6 Antioxidant material

During the manufacture and the thermal processing of a plastic material as well as its fabrications into an article it will be subjected to a variety of thermals and oxidation degradation processes which will themselves be dependent upon the nature of the polymer and its end-use. However, the oxidative degradation can be reduced by the incorporation of additives. The antioxidants help reduce the oxidation of the plastic degradation. These are dispersed or dissolved in the polymer usually by heating and melt blending. The compounds may be simple, polyfunctional or polymeric and their efficiency will depend upon a number of factors such as the temperatures used, the presence of other additives and the application or end-use, the latter case the polymer may be subjected to the extreme effects of heat, light and oxygen. In this regard antioxidant properties should be:

1.) Highly effective
2.) Colorless
3.) Non-toxic
4.) Non-volatile
5.) Stable
6.) Cost-effective

In the early years antioxidant types were divided into two main categories for inhibiting the oxidation of plastics[12]:

1. The use of what were often called “primary antioxidants” such as hindered phenols or alkylarylamines. They function by trapping or scavenging free radicals (referred to today as chain-breaking donors and/or acceptors).

2. The use of “secondary antioxidants” such as organo-sulphur or phosphorus compounds. These may be sulphides, thioethers, disulphides, mercaptans or sulphoxylates and phosphites, etc. They operate as hydroperoxide or peroxide decomposers for which the modern terminology is “peroxydolytic” antioxidants [4].

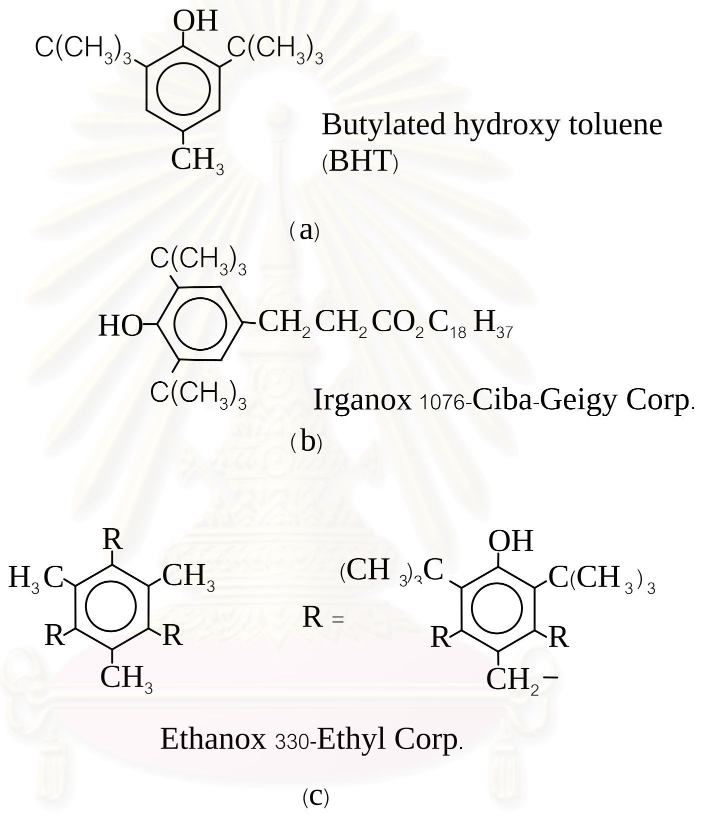
2.1.7 Antioxidant in plastics

2.1.7.1 Hindered phenolic antioxidants

These types of compounds are now well established as thermal stabilizers for a wide variety of polymer systems and today much is known about the way in which they perform. The latter will, of course, depend not only upon the structure of the stabilizer but also the nature of the polymer and conditions of degradation and/or oxidation. Some typical commercial structures are shown in Scheme 2.5, that is the simplest is 2.5 a and are widely known in the trade as simply BHT. This stands for butylated hydroxy toluene and is still widely used in a number of applications. Much of what we understand in terms of the mechanistic behavior of these types of antioxidants has originated from work on this simple structure.

The conditions of thermal treatment whether aging or processing will be quite important in terms of the nature of the mechanistic processes involved. In an enclosed environment with little or no oxygen present macroalkyl radical formation and termination will be high. We now know that the transformation products of the thermal oxidation of hindered phenols, namely the quinones, are more effective in removing macroalkyl radicals. This is called a chain-breaking acceptor mechanism (CB-A) and this works quite effectively in the polymer melt.

The second type of mechanism operates by a chain-breaking donor process (CB-D) and involves the initial phenolic hydroxyl group which is capable of donating its hydrogen atom to active free radical species in the polymer such as macroalkylperoxy radicals PO_2. This process occurs quite effectively in the presence of high concentrations of oxygen either in the melt or under low temperature aerobic ageing conditions. Thus, the ability of a hindered phenolic antioxidant to undergo a redox reaction and the relative reactivities of the oxidized and reduced forms of the molecule will be quite important in determining the stabilizing activity of the antioxidant.



Scheme 2.5 Structure of phenolic antioxidants.[9]

The general mechanism of a primary hindered phenolic antioxidant as a chain-breaking donor is shown in Scheme 2.6 [7]. Here the hydrogen atom on the phenolic hydroxy group will transfer and terminate the free radicals in the polymer produced during the thermal processing operation.

$$P\bullet + HA \longrightarrow P-H + A\bullet$$

$$PO\bullet + HA \longrightarrow POH + A\bullet$$

$$HO\bullet + HA \longrightarrow H_2O + A\bullet$$

$$PO_2\bullet + HA \longrightarrow POOH + A\bullet$$

Scheme 2.6 The general mechanism of a primary hindered phenolic antioxidant

Such radicals will then be prevented from undergoing further harmful reaction with the polymer matrix or possibly the antioxidant itself depending upon its structure and activity. This scheme is a simplistic view of the initial reaction resulting in the formation of a stabilized antioxidant radical [4].

2.1.7.2 Organophosphorus and organosulphur compounds

These are peroxide decomposers and are effective at the chain-branching stages by inducing the decomposition of hydroperoxides into inert products. By virtue of their primary function in this regard they are now termed peroxidolytic antioxidants. Consider the oxidation of cumene at 120^{o}C and the effect of a chain terminator (a) and peroxide decomposer (b) shown in Figure 2.1. Here the cumene is oxidised to a predetermined concentration of hydroperoxide (ROOH). At the inflection point on the curves for (a) the stabilizers are added. It is noted that the hindered phenol inhibits oxygen uptake initially but then after a while it continues increasing. The addition of the peroxide decomposer, diphenyl disulphide (b), inhibits the oxidation and destroys the ROOH. The disulphide is converted to a thiosulphinate and then SO_2 that is it a good peroxidize decomposer .

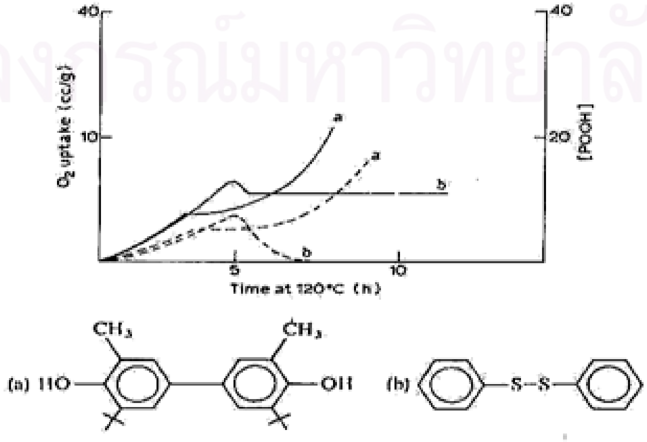


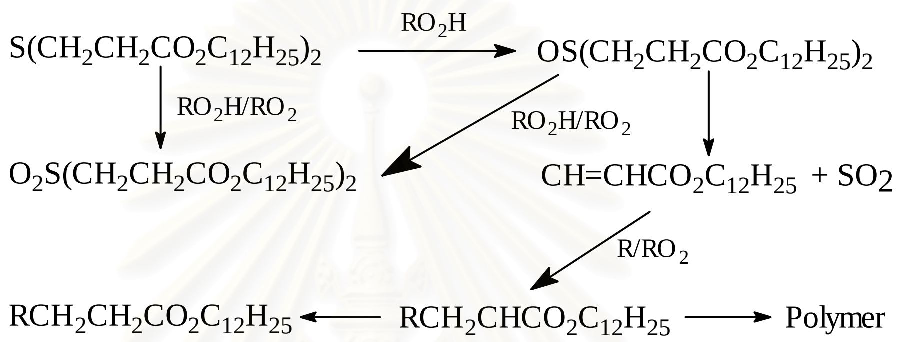
Figure 2.1 Influence of a phenolic antioxidant (a) and diphenyldisulphide (b) on the rate of oxygen uptake (___) and hydroperoxide formation (------) for cumene peroxidezed for 5 minutes at 120°C. [13]

Figure 2.4 was shown the influence of a phenolic antioxidant (a) and diphenyldisulphide (b) on the rate of oxygen uptake (-----) and hydroperoxide formation (- - -) for cumene preoxidized for 5 minutes at 120 °C. (Reproduced from F.H. Winslow, Treatise on Materials in Science and Technoloy, Vol 10, Part B. Academic Press, New York, 1977, p.741)

Sulphur and phosphorus compounds have been used in commercial practice in conjunction with hindered phenolic antioxidants as synergists. Alone they are good inhibitors of the melt degradation of polymers. This is mainly due to the primary function of the former to destroy active hydroperoxides in the melt. Aryl phosphites are well known hydroperoxide decomposers and will oxidise to phosphates. This is simplified in the Scheme 2.7 for phosphites, phosphinites and phosphines, which are subsequently oxidised to their corresponding phosphates, phosphinates and phosphine oxides. Typical sulphur containing antioxidants for polyolefins are dilaurylthiodipropionate (DLTDP) and distearylthiodipropionate (DSTDP). The mode of action of thioesters is fairly well understood with a simplified mechanistic path shown in Scheme 2.8. Here oxidation of the sulphur occurs to give a sulphoxide in the initial reaction steps. Acrylate esters and sulphur dioxide are produced, as further products which themselves will have good antioxidant behaviour.

$$\left.\begin{array}{l} \text{PHOSPHITE } P(Oph)_3 \\ \\ \text{PHOSPHINITE } PhP(Oph)_2 \\ \\ \text{PHOSPHINE } Ph_3P \end{array}\right\} \xrightarrow{RO_2H} + \begin{array}{l} \text{PHOSPHATE } OP(Oph)_3 \\ \\ \text{PHOSPHINATE } PhPO(Oph)_2 \\ \\ \text{PHOSPHINE OXIDE } Ph_3PO \end{array}$$

Scheme 2.7 Hydroperoxide reacted with phosphites [7]



Scheme 2.8 The mode action of thioesters with sulphur containing antioxidants polyolefins[7]

2.2 Literature review

A. Holmstrom and E. Sorvik [14] studied the unstabilised samples of low-density polyethylene which were heated in nitrogen containing small amounts of oxygen. The changes in molecular weight distribution were followed by gel permeation chromatography and intrinsic viscosity. A threshold temperature was found at 315^{o}C and a threshold oxygen concentration in the range 410-1000 ppm. In accordance with the theory of random scission, degradation caused a shift of peak position towards lower molecular weights and an increasing skewness of the molecular weight distributions within the original molecular weight limits. No drastic drop of M_w/M_n was observed.

S.J. Hebert and C. Tzoganakis [6] studied the properties of blown film made from post-consumer recycled (PCR) material. LLDPE film was blended with a virgin LLDPE resin at several levels in a twin screw extruder. It was shown that the interaction between recycled material content and blow-up ratio did not seem to be important. The main effect of the blow-up ratio was dominant in all properties (elmendorf tearing and dart drop impact).

S.W. Shang and R.D. Kamla [8] studied the effects of pelletizing extrusion conditions and blown film extrusion condition on the physical properties of linear low density polyethylene (LLDPE) films. The results show that better film properties can be obtained when a resin is pelletized at a lower temperature profile and then blown at a higher temperature profile. As the melt temperature of the blown film extrusion increases, the optical properties of the LLDPE film are improved. The mechanical properties, however, remain essentially the same as those blown at lower temperatures. Extrusion conditions during palletizing were shown to have a significant effect on the number of “gel” defects in film-with an increase in the melt temperature resulting in more gel.

A. Bravo and J.H. Hotchkis [5] studied the volatile compounds produced during the heating of polyethylene (PE) in the presence of excess O_2 at temperatures of 150-350^{o}C and for heating time of 5-15 min. Eighty-four volatile compounds in the range of C_5- C_{23} were identified by gas chromatography mass spectrometry. The major products were aliphatic hydrocarbons, aldhydes, ketones, and olefins. Changes in temperature and heating times affected the amount and type of compounds produced, with hexanal being found in the largest amount and 300^{o}C resulting in the greatest quantity of volatile compounds. At 350^{o}C, greater amounts and numbers of low boiling and fewer high-boiling compounds were formed. Only small amounts of volatiles were produced at 150^{o}C.

S.F. Laermer and P.F. Zambetti [12] studied the antioxidant effect of alpha-tocopherol (ATP) in biological systems, where it is better known as vitamin E, and has been well documented. However, the potential use of ATP as a stabilizer in polymers had only limited evaluation, so several studies were commissioned to obtain reliable and relevant data pertaining to the capability of ATP as a polymer additive. Data showed that ATP offers several distinct advantages in polymer stabilization. Besides its environmental safety, it is also an excellent processing stabilizer and, in combination with synergistic additives, a superior colors stabilizer. Thermal stability is excellent with no significant weight loss until almost $300^{o}C$.

P.P. Klemchuk and P.L. Horng [15] studied several quinoidal oxidation products of BHT based antioxidants which have been evaluated for UV/visible absorption characteristics in methylene chloride solutions and for contributions to Yellowness Index (YI) values in polyolefins. The quinoidal compounds with conjugated ¶-electrons exhibited the highest extinction coefficients in the visible spectrum and also contributed most of the color to polymer formulations. 3,3,’ 5,5’-Tetra-tert-butyl-stilbene 4,4’-quinone had the highest extinction coefficient in the visible spectrum and caused the highest YI values in the polyolefins.The 5 ppm presence in polyolefins was already discernible visually.

A.K. Wood, E.E. Raffy, M. Yue and G. Heally [16] studied the processing of polymers which involves prolonged shear at elevated temperature and unavoidable presence of small concentrations of oxygen. These factors can result in the degradation of the polymer. The results obtained indicated that no significant changes were occurring in the rheological and mechanical properties in the processing, even after prolonged shearing at elevated temperature, The periods of processing is well in excess of these encountered in normal processing although some slight degradation and crosslinking may be occuring.

CHAPTER III

EXPERIMENTAL PROCEDURE

3.1 Materials

3.1.1 Linear low density polyethylene (Virgin grade LLDPE)

Blown film grade linear low density polyethylene (LLDPE 1210) from Siam Polyethylene Co.,Ltd. was used. It had a melt index of 1.0 g/10 min., density of 0.919 g/cm^3, melting point of 108 ^{o}C and vicat softening point of 101 ^{o}C.

3.1.2 Waste LLDPE film

Waste LLDPE film from Prepack Thailand Co.,Ltd. was used. It was produced during the production process, mostly in the first part of new changing job which must be set for the new condition, i.e. temperature, rotating speed and blown film speed of blown film machine.

3.1.3 Polybatch AO 25

Polybatch AO 25 from PT A.Schulman plastics N.V. was used. It is an antioxidant masterbatch for the stabilization of polyethylene. Two types of antioxidant, tris(2,4-di-tert-butylphenyl)phosphite (Irgafos 168) and octadecyl 3,5-bis(1,1-dimethylethyl)-4-hydroxybenzene propanoate (Irganox 1076) were consisted in masterbatch. It had 1 part of Irganox 1076 and 2 parts of Irgafos 168 in 5.5-6.5 % of masterbatch.

Table 3.1 The properties of polybatch AO 25

Property	Units	Test Method	Limits
MFI (190°C, 2.16 kg)	G/10 min.	DIN 53735	14.0-22.0
Specific gravity	g/cm^3	DIN 63478	0.93+/-0.02
Bulk density	kg/m^3	ASTM D 1895-98	470-570
Moisture content	ppm	Schulman 456	max 1500
Base resin	-	-	PE
Pellet shape	-	-	Cylindrical
Food approval	-	-	Max 5% (BGA)
Total additive content	%	-	5.5-6.5
Pellets/Gram	p/g	Schulman 465	44-52

3.1.3.1 Irgafos 168

Irgafos 168 is an organophosphite of low volatility and particular resistance to hydrolysis. As a secondary antioxidant, it reacts during processing with hydroperoxides formed by autoxidation of organic polymers preventing process induced degradation and extending the performance of primary antioxidants and its structure is shown in Figure 3.1. It has molecular weight of 646.9 and melting point of 183-186°C in white crystalline powder form.

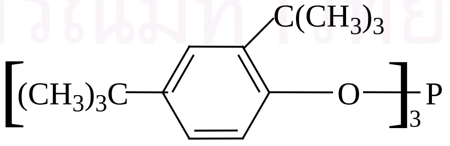


Figure 3.1 The structure of Irgafos 168[17]

3.1.3.2 Irganox 1076

Irganox 1076 is the primary antioxidant of phenolic type. It has white crystalline power form and melting point of 50-55 °C. The structure of Irganox 1076 is shown in Figure 3.2.

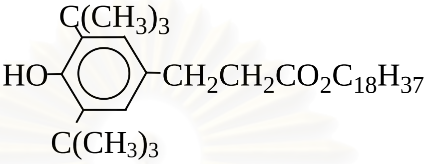


Figure 3.2 The structure of Irganox 1076 [7]

3.2 Instruments

The type of instruments used in this research are listed below.

1. Blown film extruder, TATUNG ECM-2-5 (TAIWAN)
2. Single screw extruder , SAKAI MFG (THAILAND)
3. Mixer ribbon blender, CHAOWEI KA 100 (THAILAND)
4. Granulator, CHAOWET/K 3156 (THAILAND)
5. Universal testing machine, LLOYD LS 500 (JAPAN)
6. Colorimetry, MINOLTA MODEL CR 200 (JAPAN)
7. Melt flow index testing machine, CEAST 6841/000 (ENGLAND)
8. Over head projector, 3M 2770 AHBH (TAIWAN)

3.3 Experimental procedure

3.3.1 Temperature profile in extrusion process

The temperature profile, feeding and compression zone, metering zone, filter zone and die zone in film extrusion process, was investigated with virgin grade LLDPE (LLDPE 1210) pellets. LLDPE was added in a blown film extruder with speed motor screw of 925 Hz, upper roller of 16.50 Hz and lower roller of 8.00 Hz and then performed the film. Heater temperature profiles for the blown film extruder was set at either 175°C, 190°C, 210°C, 230°C or 250°C in each batch. To observe the gel count, tensile strength and elongation were utilized for finding the appropriate temperature in each section of blown film extrusion.

3.3.2 Waste LLDPE film and recycled LLDPE film properties

The waste LLDPE film from production waste was chopped and added with AO 25. The recycled LLDPE film was prepared from waste LLDPE. Waste LLDPE was chopped from scrap LLDPE and added into a single screw extruder before blown film extrusion to form recycled LLDPE film. Waste LLDPE film and recycled LLDPE film were investigated to examine film properties, gel count, yellow index, tensile properties and tear resistance.

The blown film condition, speed motor screw of 925 Hz, air flow temperature at 40 °c, air flow rate of 2.2 m/s, upper and lower rolling speed in range of 60-95 m/min. and temperature profile followed in section 3.3.1 were used.

3.3.3 Composites preparation

3.3.3.1 Recycled LLDPE pellet preparation

The recycled pellet LLDPE was prepared from waste scrap LLDPE, waste LLDPE film after already chopped. Waste scrap LLDPE was first external blended with various loading of AO25, 0, 3, 8, 15, 25 and 40 phr in single screw extruder and then pelletized with cutting unit. These pellets were measured for melt flow index (MFI). The extruder temperature profile was set following the result of finding the temperature profile in section 3.3.1. The other conditions in blown film extrusion process are speed motor screw of 50.5 Hz,width and thickness at 280mm x 75 micron, cooling temperature in water bath at 30-35 °C and throughput 470-525 g/min. Their formulations are given in Table 3.1.

Table 3.2 Formulations of recycled pellet LLDPE in single screw extruder.

Formular No.	Waste scrape LLDPE (pbw)	AO25 (phr)
P0	100	0
P3	100	3
P8	100	8
P15	100	15
P25	100	25
P40	100	40

3.3.3.2 Recycled LLDPE film preparation

The recycled LLDPE pellet which was prepared from section 3.3.3.1 was external blended with various virgin grade LLDPE loading at 0, 25, 50, 75 and 100 pbw in mixer ribbon blender. These blends were ultimately molded with blown film extrusion. The extruder temperature profile was set following the result of finding the temperature profile in section 3.3.1. The other condition in blown film extrusion process, speed motor screw of 925 Hz, air flow temperature at 40^{o}C, air flow rate of 2.2 m/s and upper and haul off rolling speed of 6.0-9.5 m/min., were used. Their formulations are given in Table 3.2.

Each film composition from blown film extrusion was measured for the gel count, yellowness index (YI), tensile property and tear resistance, sections 3.5-3.8, respectively.

Table 3.3 Formulations of recycled LLDPE film in blown film extruder.

Recycled LLDPE film No.	Recycled pellet LLDPE (pbw)	Virgin grade LLDPE (pbw)
Virgin LLDPE film	0	100
P0/25	25	75
P0/50	50	50
P0/75	75	25
P0/100	100	0
P3/25	25	75
P3/50	50	50
P3/75	75	25
P3/100	100	0
P8/25	25	75
P8/50	50	50
P8/75	75	25
P8/100	100	0
P15/25	25	75
P15/50	50	50
P15/75	75	25
P15/100	100	0
P25/25	25	75
P25/50	50	50
P25/75	75	25
P25/100	100	0
P40/25	25	75
P40/50	50	50
P40/75	75	25
P40/100	100	0

3.4 Melt flow index (MFI)

The measuring melt flow index (MFI) was carried out according to ASTM D 1238 [18] by melt flow index testing machine, ceast 6841/000 model. The pellet samples, each composition of waste scrap LLDPE/AO 25 blend from single screw extrusion process before molded with blown film extrusion process, of 3-4 gram were heated at 190^{o}C in extrusion plastometer and were compressed with 2.16 kg dead load. The sample weight flew out was recorded for 10 minutes.

3.5 Measuring of Gel count

This test method covers the determination of the number of gels present in plastic film when observing the images formed on a screen when light is projected through the film according to ASTM D 3351-74 [19]. It was developed because the counting of gels in plastic film with the unaided eye is difficult and fatiguing to the operator.

A 3M model 2770 overhead projector was used to magnify the film surface ten times (10x) for better inspection. The gel counts were averaged from 4 strips of 8 in. x 8 in. cut from 4 different sections of the blown films. The 4 strips blown films were placed on the overhead projector, bring the acrylic plate putted on these films and press firmly to remove wrinkles as far as possible. Focus the projector so that the gel images and the image of the gel size are clearly projected on the screen and outcome of this testing was a total and individual size range gel count per 1520 cm^2 of testing area. The blown film appearance defects were counted and classified into three different size categories, 0.4 to 0.8 mm, 0.8 to 1.5 mm and greater than 1.5 mm.

Acrylic plate size of 9.0 by 9.0 by 3.2 in. was contained three standard holes, 0.4, 0.8 and 1.5 mm. in diameter and drawn rectangle of 19.0 by 20.0 cm. to form the cover area of these standard holes.

3.6 Measuring of Yellowness index (YI)

The Yellownes Index (YI) was determined according to ASTM D 1925-70[20] on a Minolta Colorimeter Model CR 200.

The five tube shaped-testing film were stacked and put on A4 paper before illuminated under day-light with CR 200. This machine will measure the YI value. The comparison of YI between testing film should be made only for the same film thickness because the YI of transparent and translucent plastics is a function of thickness.

3.7 Measuring of Tensile property

The tensile test specimens of film from the blown film extruder with dimensions of 10 mm. by 100 mm. by 0.075 mm. were measured using Universal Testing Machine (LLOYD LS 500) following the procedures described in ASTM D 882-91[21].

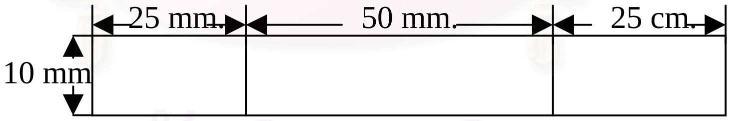


Figure 3.3 Film dimensions of tensile test specimen.

The tensile parameters and testing condition, speeding testing of 500 mm/min, gauge length of 50 mm. and load cell of 50.99 kg_f were used. The tensile strength at break (kg_f/mm^2) and elongation at break (%) were recorded.

3.8 Measuring of Tear resistance

The tear resistance method is the determination of force necessary to propagate a tear in plastic film by Universal Testing Machine (LLOYD LS 500), according to ASTM D 1938-92 [22]. The dimensions of single-tear specimen are shown in Figure 3.4. It consists of strips of 75 mm. long, by 25 mm. wide, longitudinal slit of 50 mm. long cut with a sharp razor blade and thickness 0.075 ± 0.002 mm.

The single-tear testing condition, speeding testing of 250 mm/min, gauge length of 50 mm., load cell of 50.99 kg_f and temperature at 23 ± 2°C, were used. The initial force to continue the propagation of the slit and the maximum force were reported in kg_f unit.

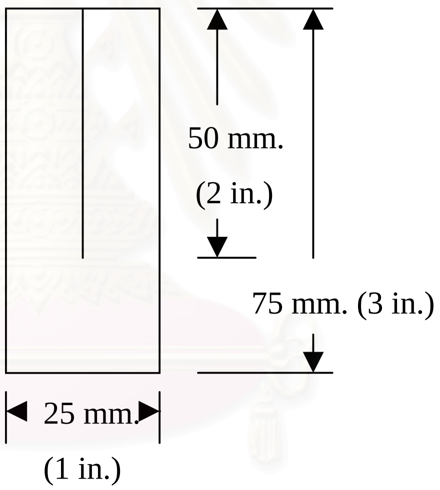


Figure 3.4 Tear resistance specimen

CHAPTER IV

RESULTS AND DISCUSSION

4.1 Temperature profile in extrusion process

Extrusion processability and subsequent film properties are two critical elements used by film extrusion process in selecting a specific resin for their applications. The blown film processing condition which one factor affecting extrusion, the temperature in each zone of extruder, was the first study with virgin grade LLDPE in this work. The effects of extrusion temperature on gel count, tensile strength and elongation are shown in Table 4.1(see Appendix A and B). The speed of upper and lower roller in blown film extrusion were set at 16.50 and 8.00 rpm, respectively.

Table 4.1 Effect of temperature in blown film extrusion on virgin grade LLDPE film properties.

Temperature profile (^{o}C)		**175**	**190**	**210**	**230**	**250**
Ultimate tensile strength at break. (kg_f / mm^2)	MD	3.04	3.37	3.27	3.12	2.73
	TD	2.73	2.90	2.95	2.78	2.30
Ultimate elongation at break. (%)	MD	946	1072	1148	1072	869
	TD	1172	1187	1207	1087	977
Gel counts:						
0.4-0.8 mm.		45	20	18	32	31
0.8-1.5 mm.		7	6	10	9	10
> 1.5 mm.		2	1	3	1	3
Total		**54**	**27**	**31**	**42**	**44**

From Table 4.1, it can be observed that the ultimate tensile strength both of MD and TD is continuously increased until the temperature profile is 210°C and decreased as the temperature profile is higher than 210°C. However, the temperature in the range of 190-210°C gives the best result in tensile strength and also elongation. Total gel counts are decreased until the temperature is at 210°C and increased after this.

The results of most tensile strength, elongation and gel counts of virgin grade LLDPE with various temperature profile indicated that the optimum temperature profile for extrusion process is 190-210°C. Consequently, the process for preparation of pellet LLDPE and recycled LLDPE in this work was set for the screw temperature in feeding and compression zone, metering zone and die zone at 190, 195, 205 and 210°C, respectively, as shown in Table 4.2.

Table 4.2 Single screw and blown film extrusion process

Extrusion process	**Temperature profile of extrusion zone (°C)**			**Die zone**
	Feeding and Compression	**Metering**	**Filter**	
Single screw extrusion	190	200	-	210
Blown film extrusion	190	195	205	210

4.2 Waste LLDPE film and recycled LLDPE (P0/100) properties.

In this section, waste LLDPE films from production waste before choped and recycled LLDPE (P0/100) were investigated to examine film properties, tensile properties, tear resistance, yellowness index (YI) and gel counts. Those properties are shown in Table 4.3 (see Appendix A,B,C and E).

Table 4.3 Waste LLDPE and recycled LLDPE film properties.

Film properties		Waste LLDPE film	Recycled LLDPE film
Ult.tensilestrength at break (kg$_f$ / mm^2)	MD	2.28	2.18
	TD	2.15	1.97
Ult.elongation At break (%)	MD	747	676
	TD	950	824
Tear resistance (kg$_f$)	MD	0.81	0.68
	TD	0.75	0.66
Yellowness Index (YI)		-9.07	-7.06
Gel counts:			
0.4– 0.8 mm.		4	9
0.8–1.5 mm.		0	3
> 1.5 mm.		0	2
Total		**4**	**14**

From Table 4.3, when waste LLDPE film was extruded again to form recycled LLDPE film all the physical properties were lower than those of the original waste LLDPE film. The recycled LLDPE film had the highest gel levels and YI. This result caused by the occurring of oxidation reaction in extrusion process once again after production process. In that way, the improvement of the recycled LLDPE film properties can be reduced by the addition of antioxidant during extrusion process.

4.3 Processability of recycled pellet LLDPE

The melt flow index (MFI) of the blends of waste scrap LLDPE filled with various amount of AO25 were measured to observe the processability in extrusion process. These blends were measured after single screw extrusion process. The data are shown in Table 4.4 (see Appendix E).

Table 4.4 MFI of recycled pellet LLDPE

Composite Number.	**Waste scrap LLDPE (pbw)**	**AO 25 (phr)**	**Melt flow index : MFI (g/10 min.)**
P0	100	0	1.02
P3	100	3	1.08
P8	100	8	1.23
P15	100	15	1.59
P25	100	25	1.77
P40	100	40	2.13

From the above result, it was observed that the MFI is higher when the amount of AO 25 is increased. This result is due to the characteristic of AO25 which has higher MFI (14-22 g/min.). The higher value of MFI effected the blown film extrusion process so that the MFI value is an important parameter. In generally, blown film extrusion of LLDPE should be in the range of 0.2-2.2 see Figure 4.1. However, the MFI of all recycled pellets LLDPE in this work is 1.02-2.13 g/10 min. Consequently, it can be concluded that recycled pellet LLDPE can be processed in blown film extrusion.

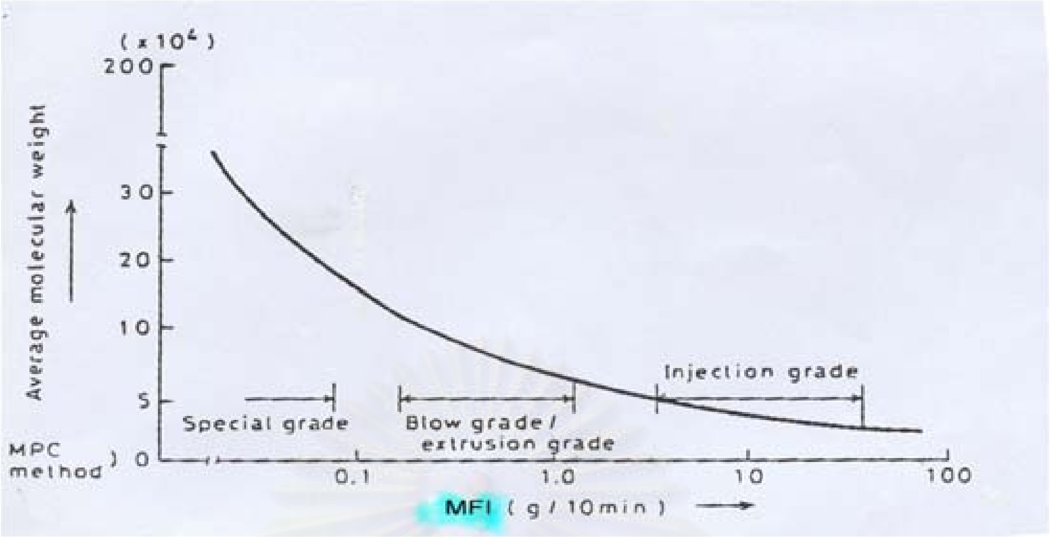


Figure 4.1 Relation of MFI and extrution process for LLDPE [23]

4.4 Recycled LLDPE film

In this section, the oxidation reaction which produced gel formation in recycled LLDPE caused the drop in film properties, e.g. tensile strength and elongation. The film properties of recycled LLDPE film, various amount of recycled pellet LLDPE and virgin grade LLDPE blends, are shown in Sections 4.4.1-4.4.4.

4.4.1 Gel counts of recycled LLDPE film

Gel formation of film caused by many type, e.g. contamination from dirt, unmelted of polymer and polymer degradation gel from high crosslinking of polymer and oxidation reaction in the presence of oxygen. However, the purpose in this study is focus on the reduction of the oxidation reaction by adding AO25. Moreover, the virgin grade LLDPE was used to reduce gel count as well.

The effect of amount of virgin grade LLDPE and AO25, on gel counts of recycled LLDPE film is shown in Table 4.5. The total gel counts of all recycled LLDPE film are shown in Figure 4.2.

Table 4.5 The effect of the amount of virgin grade LLDPE and AO25 on gel counts of recycled LLDPE film.

Film sample	**Gel levels**			
	0.4-0.8 mm.	**0.8-1.5 mm.**	**> 1.5 mm.**	**Total**
Virgin LLDPE film	3	1	0	**4**
P0/25	7	5	0	**12**
P0/50	9	5	0	**14**
P0/75	10	8	0	**18**
P0/100	12	6	2	**20**
P3/25	2	6	0	**8**
P3/50	5	4	1	**10**
P3/75	11	2	0	**13**
P3/100	12	2	1	**15**
P8/25	4	2	0	**6**
P8/50	8	2	0	**10**
P8/75	7	4	1	**12**
P8/100	8	2	2	**12**
P15/25	4	2	0	**6**
P15/50	3	3	0	**6**
P15/75	8	1	0	**9**
P15/100	8	0	1	**9**
P25/25	2	4	0	**6**
P25/50	2	5	0	**7**
P25/75	7	2	0	**9**
P25/100	4	5	0	**9**
P40/25	3	2	1	**6**
P40/50	3	4	0	**7**
P40/75	3	5	0	**8**
P40/100	4	3	1	**8**

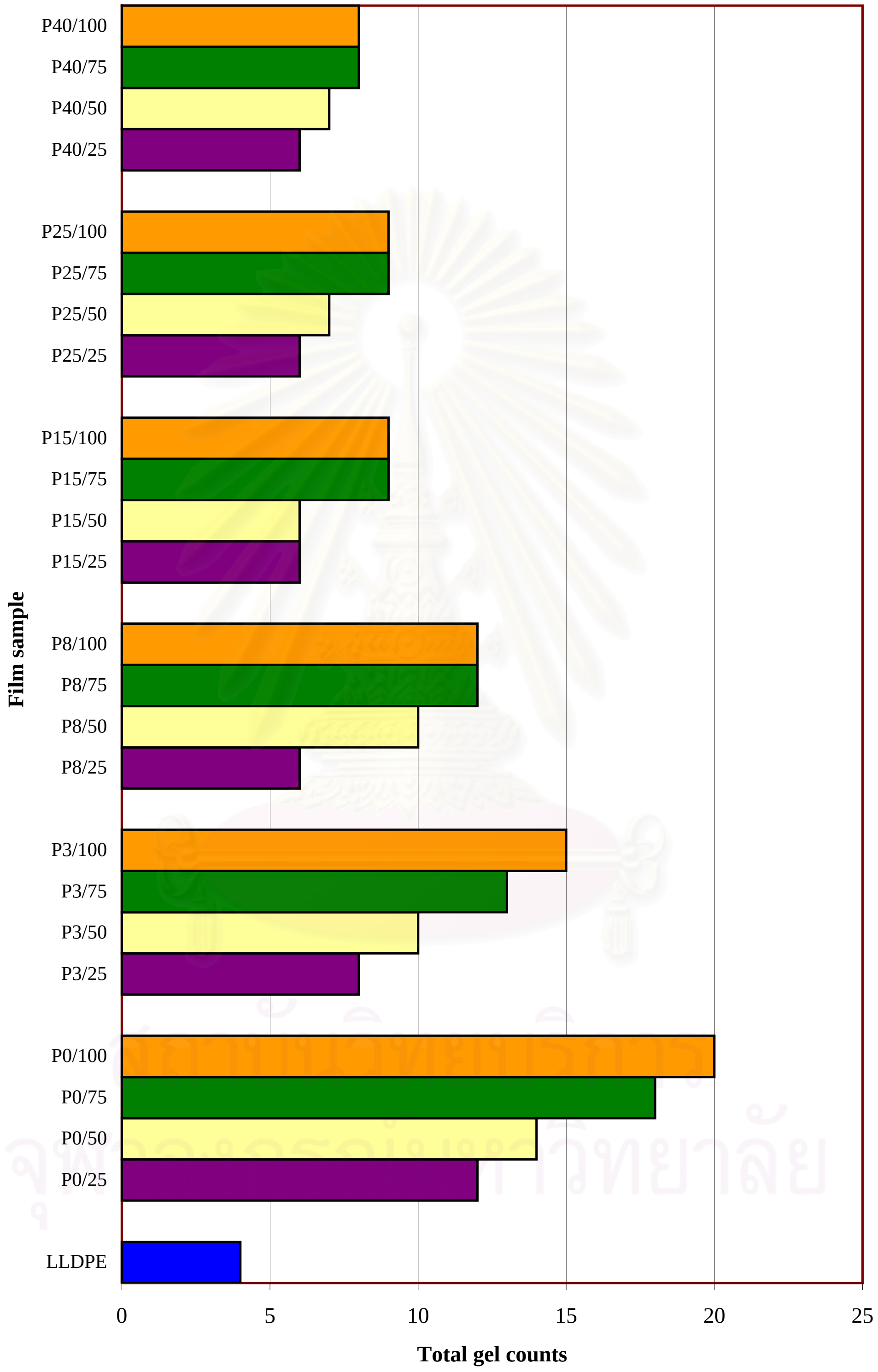
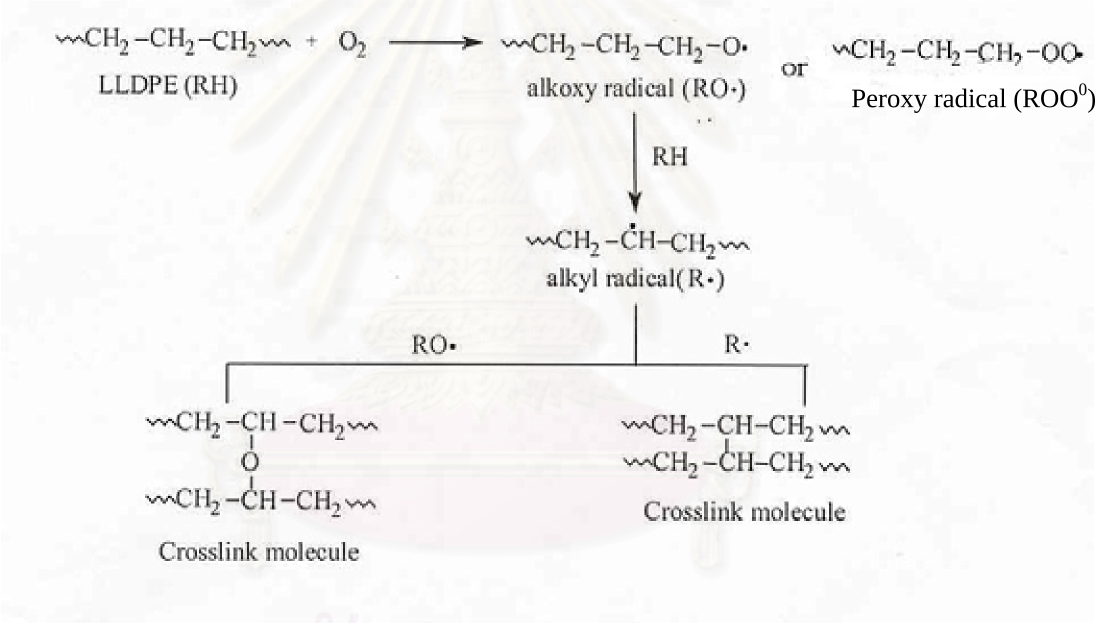


Figure 4.2 Gel counts at various contents of recycled pellet and virgin grade LLDPE.

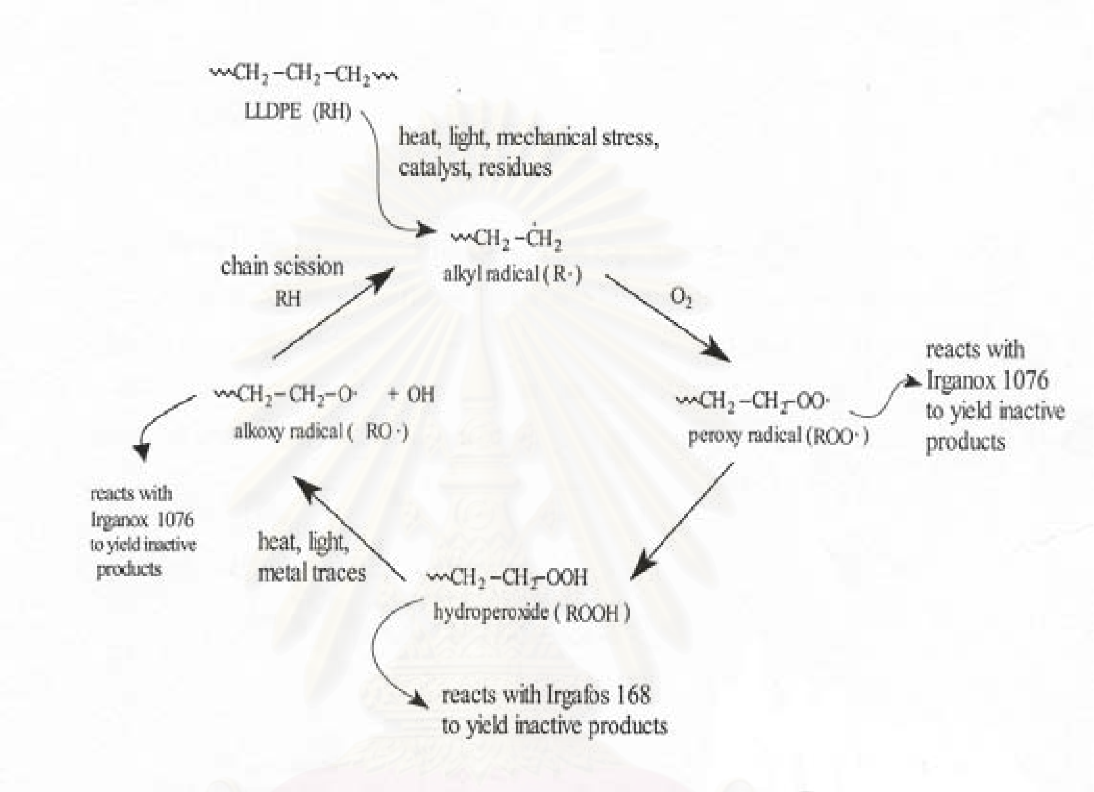
From Figure 4.2, it was observed that gel counts varied with the amount of antioxidant. When higher antioxidant was added the gel counts are lower, comparing to the composition that have the same recycled pellet LLDPE adding, e.g. P0/25, P3/25, P8/25, P15/25, P25/25 and P40/25. However, gel counts are rather constant when more than 15 phr antioxidant was added.

One factor effecting gel formation is the atmospheric oxygen. It can react with LLDPE to generate alkoxy radical (RO•) and peroxy radical (ROO•) which can propagate the reaction with LLDPE to form alkyl radical (R•). This alkyl radical will reactively combined with a R• or RO• to form a crosslinking higher molecular weight compound (oxidized gel), see Scheme 4.1.

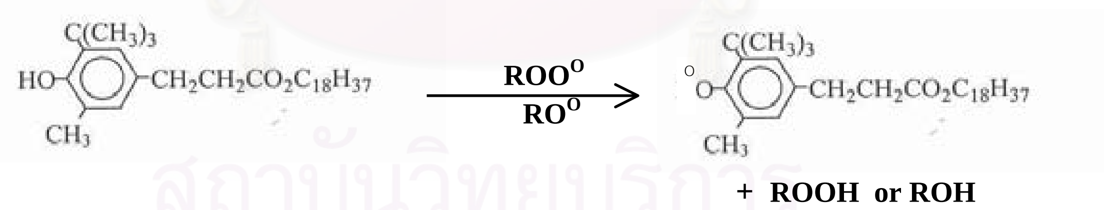


Scheme 4.1 The oxidized gel reaction in LLDPE [24]

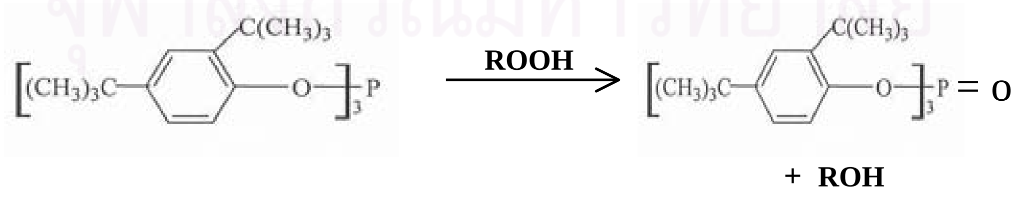
In general, gel formation from crosslinking of thermoplastic can be reduced by the addition of antioxidant. In this work, Irganox 1076 and Irgafos 168 were used to react with peroxy radicals (ROO•), alkoxy radical (RO•) and hydroperoxide (ROOH), respectively, see Scheme 4.2. The ROO• is generated from the reaction between R• and atmorpheric oxygen (O_2) and it will reactively combined with Irganox 1076, a primary antioxidant, to yield inactive products. At the same time the ROO• that can not react with Irganox 1076 will thus reactively combined with LLDPE to form hydroperoxide (ROOH). It is able to react with Irgafos 168, a secondary antioxidant, to yield inactive products too. But, there is some ROOH that can not react with Irgafos 168 and it will generate the alkoxy radical(RO•) and they can react with Irganox 1076 to terminate active RO•.



a) The antioxidations reaction process



b) Irganox 1076 reaction



c) Irgafos 168 reaction

Scheme 4.2 LLDPE degradation and reaction of primary and secondary antioxidant [25].

From above results, it can be concluded that gel formation of recycled LLDPE film was reduced by the addition of antioxidant. However, more antioxidant (more than 15 phr-40 phr) was not useful in reducing gel counts.

Considering of the amount of virgin grade LLDPE was decreased that has same amount of AO 25, are shown the increasing in gel counts. This is because recycled pellet LLDPE was more oxidized than virgin grade LLDPE. Moreover, it was already processed. For instance, the composition number P3/25 exhibited gel counts at 8 which was lower than that of P3/75 (13 gels).

From all the results, it can be concluded that the two factors for decreasing the gel counts of recycled LLDPE film are AO25 and virgin grade LLDPE. In addition, P8/25, P15/25, P15/50, P25/25 and P40/25 show the lowest in gel counts (6 gels) which are almost similar of virgin grade LLDPE film (4 gels). However, P8/25 is seen as the best composition for the application in industry due to the lowest AO 25 use, which is the best cost effective.

4.4.2 Tensile properties of recycled LLDPE film

The tensile property, tensile strength at break and elongation at break, of recycled LLDPE film are shown in Table 4.6(see Appendix B). Figure 4.3 (a) and (b) were shown tensile strength at break at different direction, MD and TD, respectively. Figure 4.4 (a) and (b) illustrate the elongation at break at MD and TD, respectively.

From Figures 4.3 and 4.4, it was observed that the amount of AO25 and virgin grade LLDPE or recycled pellet LLDPE effected the tensile strength at break and elongation at break. When considering the film sample that the increasing of AO25 content with fix amount of recycled pellet LLDPE, e.g. P0/25 and P3/25, the enhancing in tensile strength at break and elongation at break was observed. However, this result is shown by the addition of AO 25 at only 0-3 phr. These tensile properties was continuously dropped by adding more AO25, e.g. P8/50 exhibited 2.44 kg_f/mm^2 of tensile strength at break (MD) and 888% elongation at break (MD) whereas P40/50 exhibited 1.60 kg_f/mm^2 and 664%. Comparing these sample films, the tensile strength at break and elongation at break were significantly decreased to 52.5% and 34% from P8/50 to P40/50, respectively.

In case of adding AO 25 from 8 phr up to 40 phr, the decreasing in tensile property was explained that the increasing MFI of recycled pellet LLDPE was caused by adding more AO 25 that has MFI up to 14-22 g/10 min.(see Figure 3.1). This indicated that the average molecular weight was low (see Figure 4.1). In generally, the tensile properties related to MFI converting, see Figure 4.5.

Table 4.6 Tensile properties of recycled LLDPE film

Film sample	**Tensile strength at break (kg_f/mm^2)**		**Elongation at break (%)**	
	MD	**TD**	**MD**	**TD**
Virgin LLDPE film	3.01	2.99	1051	1057
P0/25	2.90	2.39	937	1027
P0/50	2.55	2.15	909	1009
P0/75	2.25	2.01	849	950
P0/100	2.18	1.99	788	932
P3/25	2.94	2.54	937	1056
P3/50	2.68	2.25	909	1013
P3/75	2.29	2.07	852	955
P3/100	2.23	2.00	794	947
P8/25	2.84	2.38	911	1014
P8/50	2.44	2.09	888	971
P8/75	2.18	1.98	782	928
P8/100	2.04	1.83	756	862
P15/25	2.15	1.98	777	920
P15/50	2.01	1.92	758	874
P15/75	2.04	1.85	750	833
P15/100	1.85	1.90	750	828
P25/25	2.13	1.93	760	895
P25/50	2.01	1.79	748	825
P25/75	1.98	1.68	741	804
P25/100	1.99	1.73	742	824
P40/25	1.82	1.66	734	801
P40/50	1.60	1.61	664	785
P40/75	1.64	1.63	677	798
P40/100	1.53	1.29	616	743

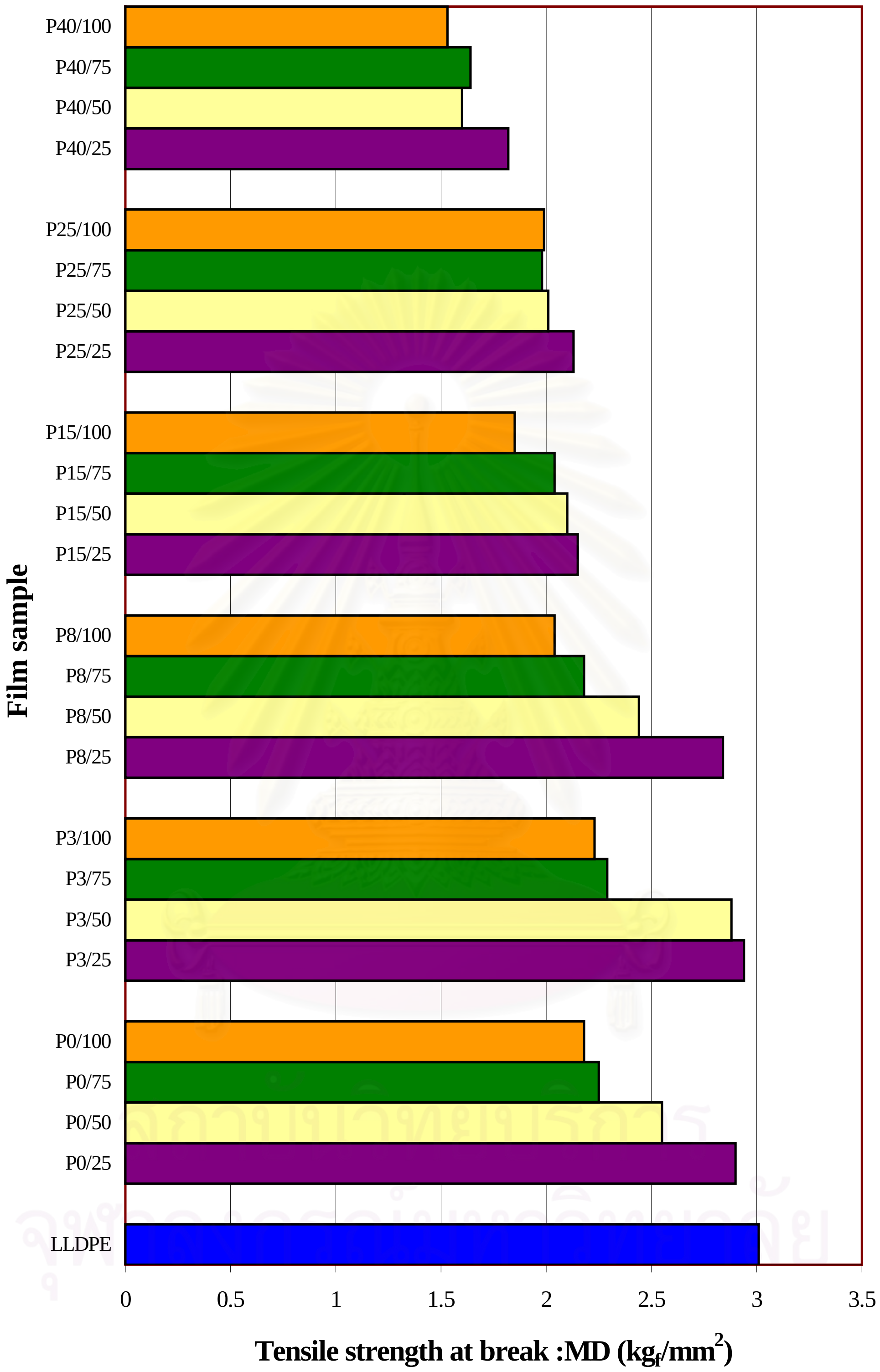


Figure 4.3 (a) Tensile strength at break (MD) at various contents of recycled pellet and virgin grade LLDPE.

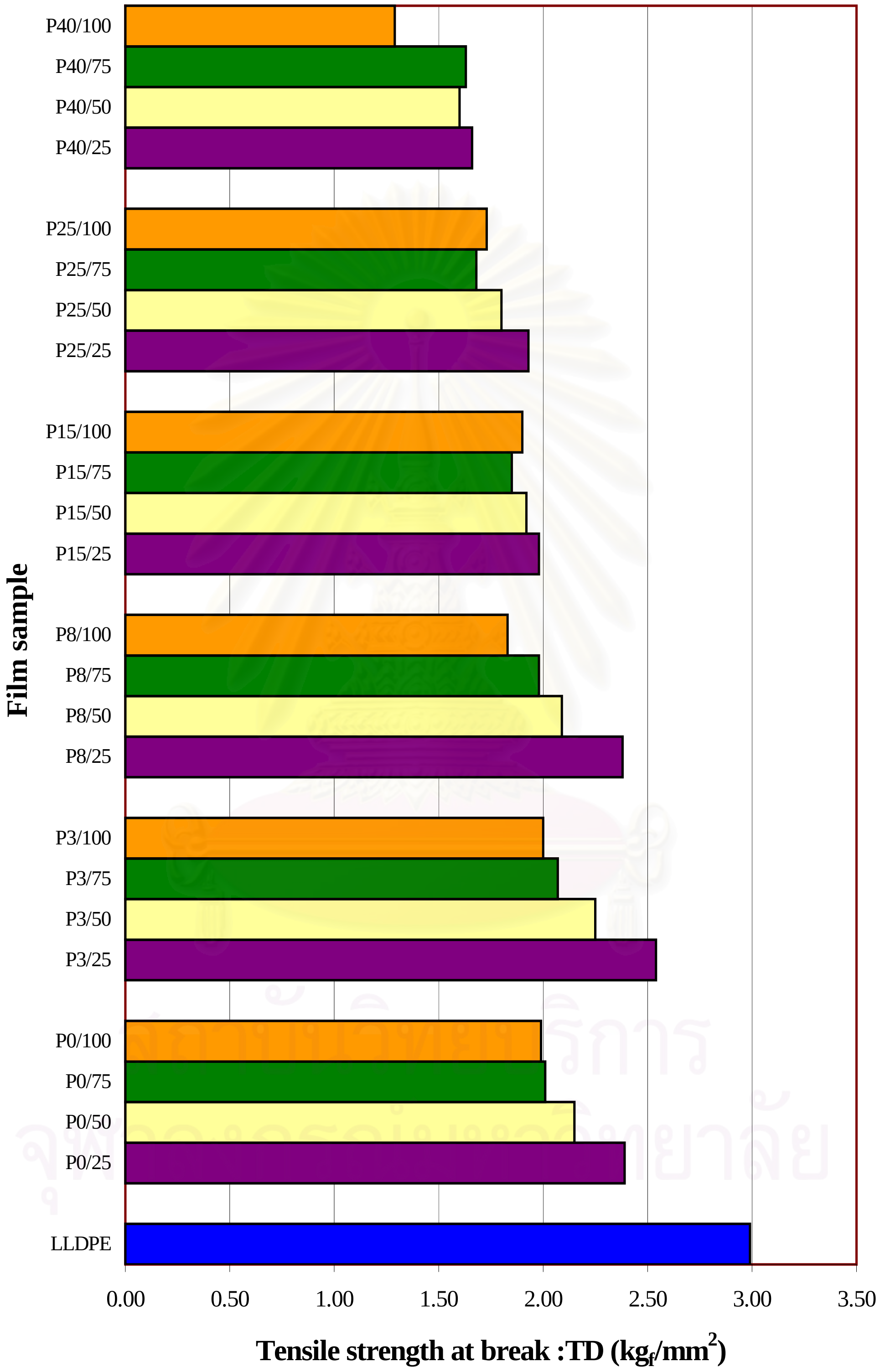


Figure 4.3 (b) Tensile strength at break (TD) at various contents of recycled pellet and virgin grade LLDPE.

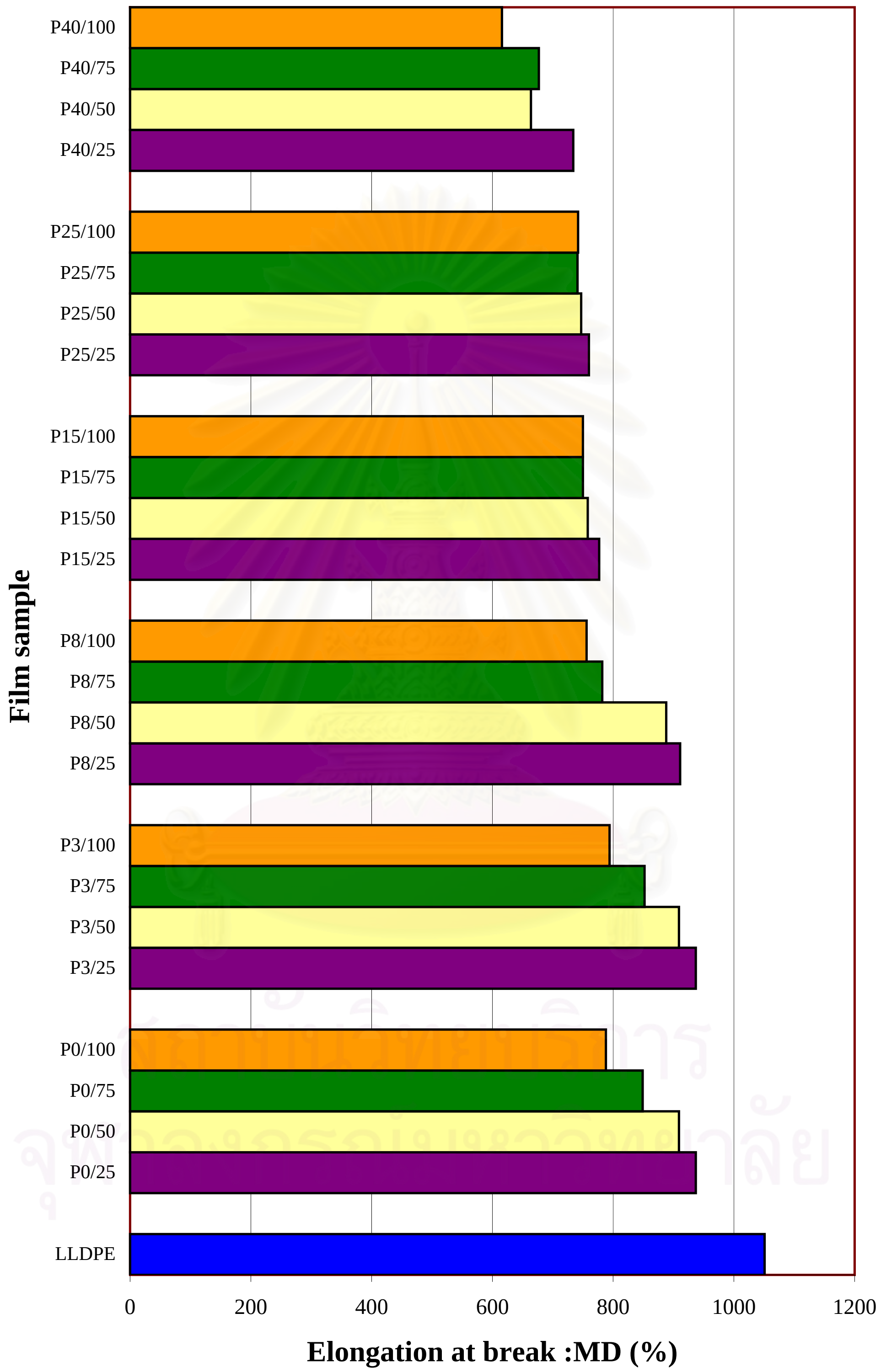


Figure 4.4 (a) Elongation at break (MD) at various contents of recycled pellet and virgin grade LLDPE.

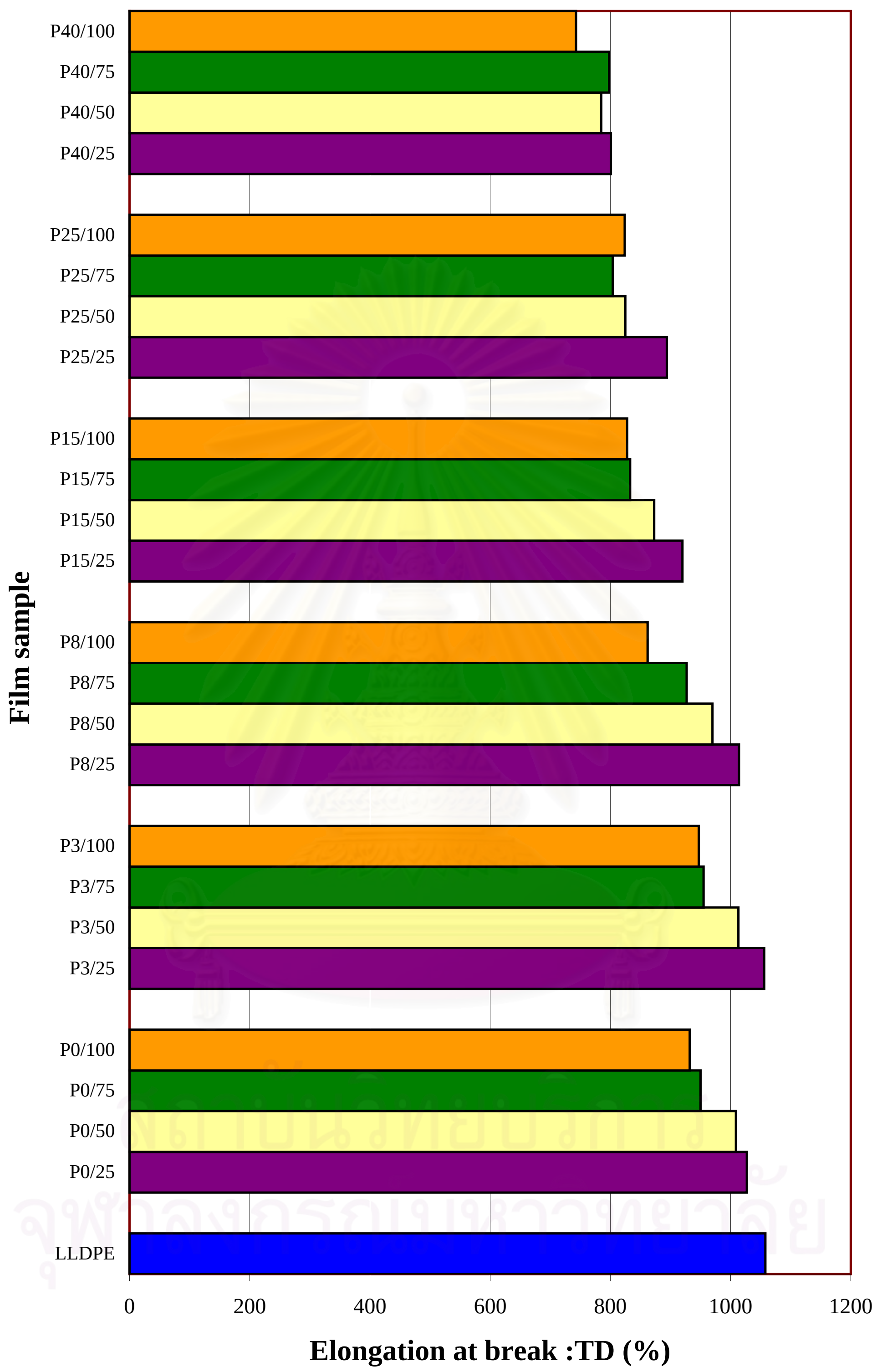


Figure 4.4 (b) Elongation at break (TD) at various contents of recycled pellet and virgin grade LLDPE.

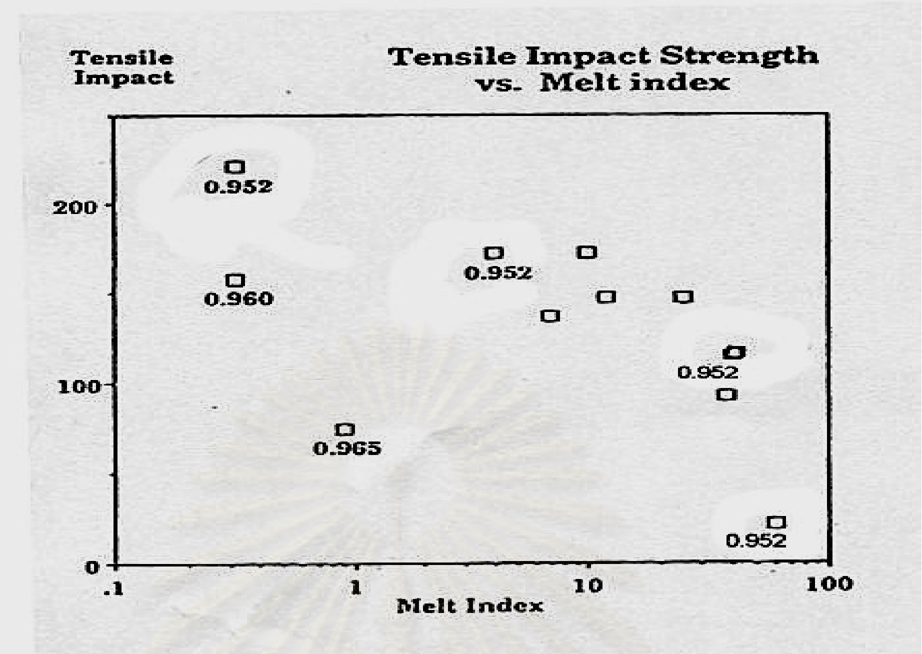


Figure 4.5 Relationship of MFI and tensile properties [26]

The enhancing tensile properties, in case of adding 0-3 phr AO25 in recycled LLDPE film, resulted from the chain scission generation in LLDPE chain which caused the drop in MFI .

It was also observed that tensile properties continuously decreased (increased MFI) when increasing amount of recycled pellet LLDPE at added constant of AO 25. For example, comparing sample P15/25 with P15/100 showed the tensile strength at break (MD) of 2.15 and 1.85 kg_f/mm^2, respectively and elongation at break (MD) of 777 and 750 %, respectively. The recycled LLDPE film number P15/25 was shown tensile properties more than P15/100 due to more chain scission formation of recycled LLDPE film that more recycled pellet LLDPE adding. The recycled pellet LLDPE has opportunity to form chain scission more than virgin grade LLDPE.

4.4.3 Tear resistance of recycled LLDPE film

Table 4.7(see Appendix C) shows tear resistance data both MD and TD direction of recycled LLDPE film. Figure 4.6 shows the plot of film sample and tear resistance.

Table 4.7 Tear resistance of recycled LLDPE film

Film sample	**Tear resistance (kg_f)**	
	MD	**TD**
Virgin LLDPE film	0.90	0.89
P0/25	0.83	0.88
P0/50	0.80	0.85
P0/75	0.78	0.85
P0/100	0.78	0.82
P3/25	0.83	0.88
P3/50	0.81	0.87
P3/75	0.79	0.84
P3/100	0.78	0.83
P8/25	0.82	0.87
P8/50	0.80	0.85
P8/75	0.78	0.82
P8/100	0.74	0.80
P15/25	0.76	0.81
P15/50	0.75	0.80
P15/75	0.72	0.79
P15/100	0.71	0.76
P25/25	0.76	0.80
P25/50	0.71	0.78
P25/75	0.70	0.77
P25/100	0.70	0.78
P40/25	0.69	0.70
P40/50	0.62	0.66
P40/75	0.63	0.69
P40/100	0.59	0.63

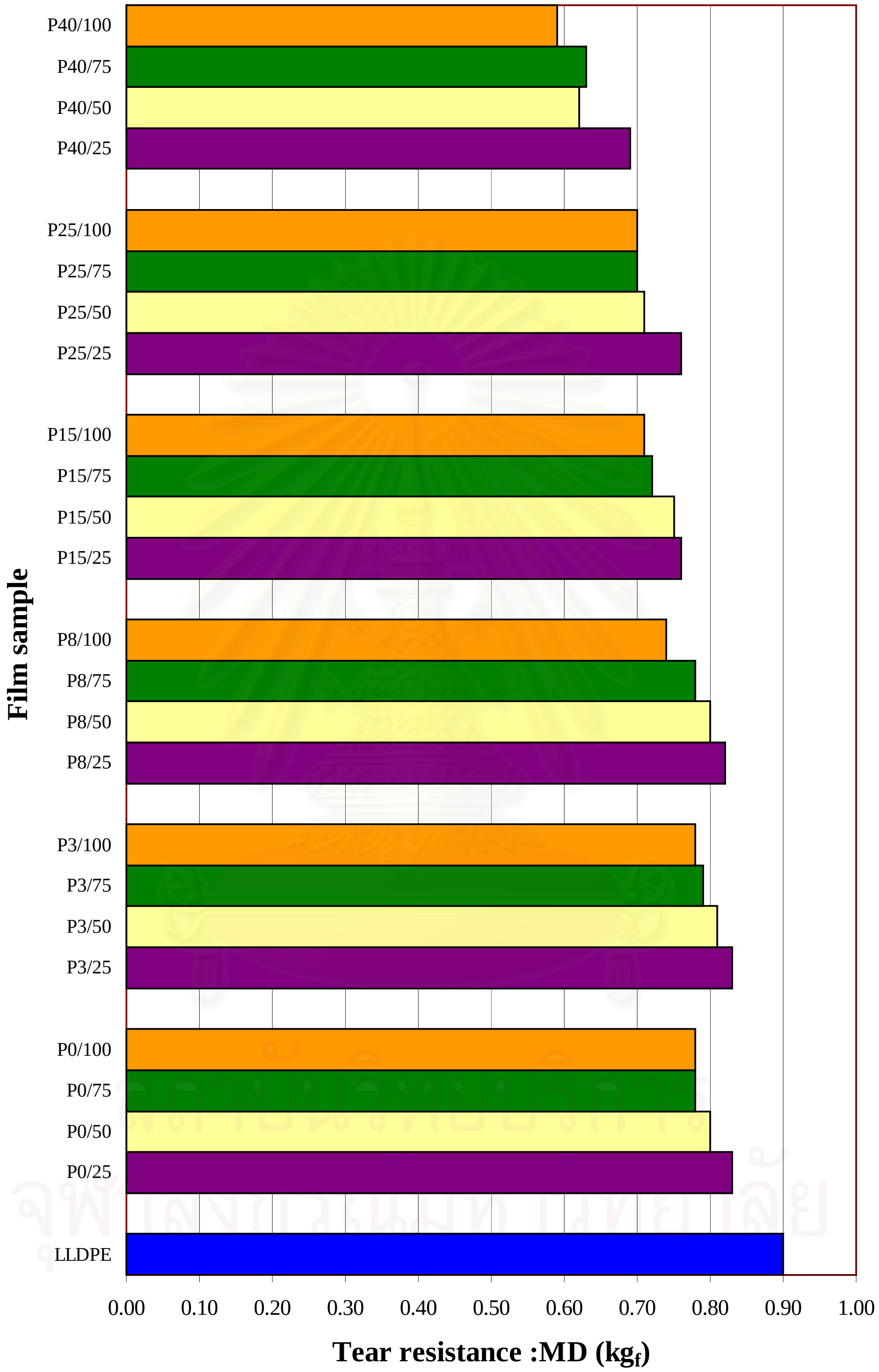


Figure 4.6 (a) Tear resistance (MD) at various contents of recycled pellet and virgin grade LLDPE.

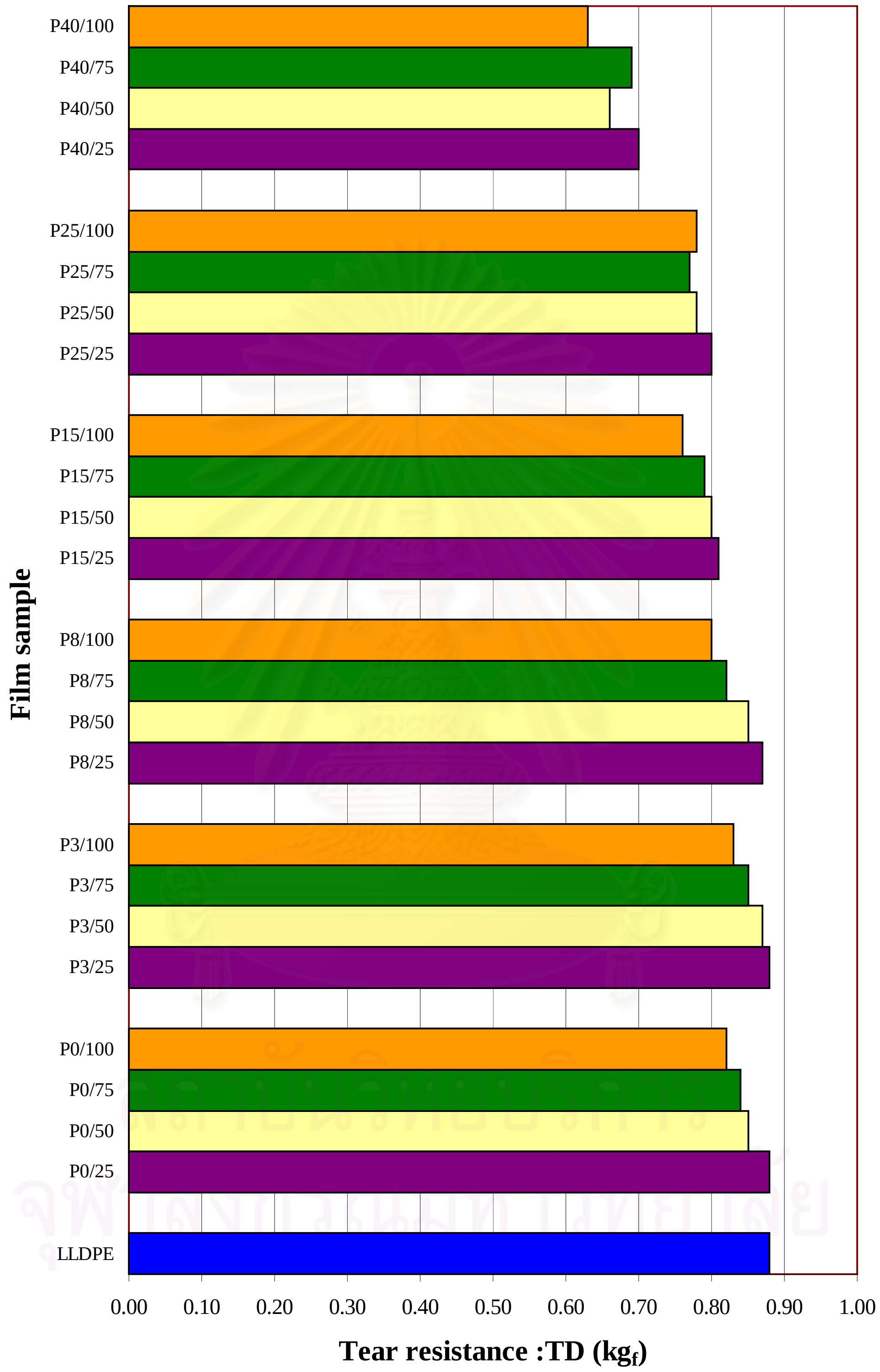


Figure 4.6 (b) Tear resistance (TD) at various contents of recycled pellet and virgin grade LLDPE.

The tear resistannce of recycled LLDPE film was similar to tensile properties. Thus, the amount of AO25 and recycled pellet LLDPE or virgin grade LLDPE effected to tear resistance of these films. The tear resistance of recycled LLDPE film that has adding AO25 from 0 to 3 phr. was almost the same result. However, it was continuously decreasing by adding AO25 more than 3 phr, comparing with constant of recycled pellet LLDPE.

In case of adding AO 25 from 8 phr up to 40 phr, the decreasing in tear resistance properties was explained that the increasing MFI of recycled pellet LLDPE was caused by adding more amount of AO 25 that has MFI up to 14-22 g/10 min.(see Figure 3.1). This indicated that the average molecular weight is low. In case of adding AO25 into recycled LLDPE film in range at 0-3 phr could be caused MFI drop which has resulted from the chain scission generation in LLDPE chain.

4.4.4 Yellowness index of recycled LLDPE film

The color of recycled LLDPE film was measured by the yellowness index (YI) as suggested by UV absorption in the visible. The data in Table 4.8(see appendix D) shows the YI of recycled LLDPE film. Figure 4.7 shows the plot of YI versus the film sample of recycled LLDPE film.

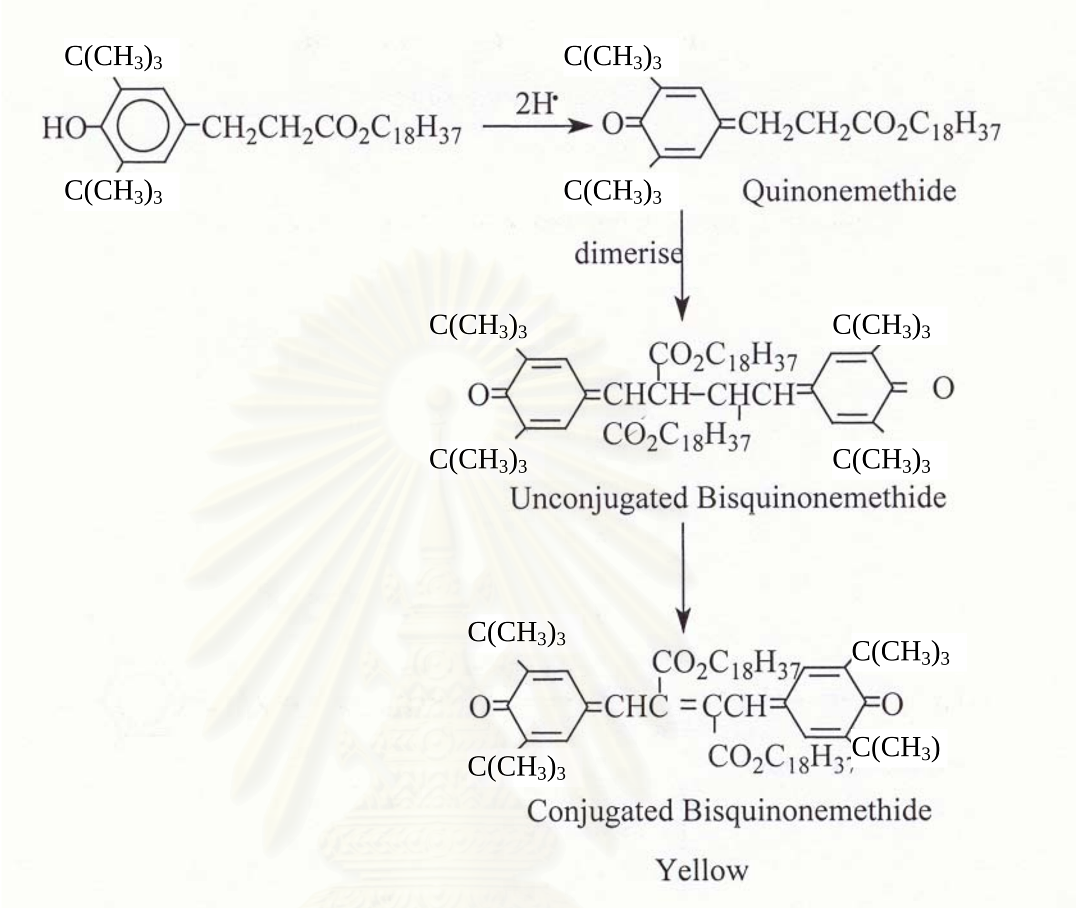
It can be observed that the P0/100 film has higher value (-7.24) than virgin LLDPE film (-7.72), see Figure 4.7. It is also indicated that P0/100 film has higher yellow color than virgin LLDPE film. In addition the higher adding of AO25 into recycled LLDPE film was observed the higher yellowness.

The above results caused by transformation reaction of Irganox 1076 which commonly used for the stabilization of LLDPE against to oxidation reaction, see Scheme 4.3.

Table 4.8 Yellowness index (YI) of recycled LLDPE film

Film sample	Yellowness index : YI
Virgin LLDPE film	-7.72
P0/25	-7.54
P0/50	-7.48
P0/75	-7.29
P0/100	-7.24
P3/25	-7.58
P3/50	-7.35
P3/75	-7.22
P3/100	-6.96
P8/25	-7.47
P8/50	-6.90
P8/75	-6.70
P8/100	-6.42
P15/25	-7.00
P15/50	-6.85
P15/75	-6.62
P15/100	-6.41
P25/25	-6.80
P25/50	-6.21
P25/75	-6.44
P25/100	-5.87
P40/25	-6.60
P40/50	-6.29
P40/75	-6.02
P40/100	-5.82

Figure 4.7 Yellowness index at various contents of recycled pellet and virgin grade LLDPE.



Scheme 4.3 Yellowing film reaction from Irganox 1076 [27]

Yellowing of film development in recycled LLDPE film, caused by nitrogen oxide from exhaustion, polymer catalyst residues, pigments is attributed to unconjugated bisquinonemethide and conjugated bisquinonemethide transformation products resulting from the trapping of oxygen-centred radical to terminate reactions. However, the conjugated bisquinonemethide contributed more color to the LLDPE than the unconjugated bisquinonemethide. Thus, the higher AO25 added, the higher transformation to unconjucated compound has been observed.

However, Irgafos 168 can be transformed neither unconjugated bisquinonemethide nor conjugated bisquinonemethide causes yellowing film .

The addition of AO25 can reduce the gel formation. But, enhance the YI. Thus P8/25 was the best recycled LLDPE film due to the least of adding AO25.

CHAPTER V

CONCLUSION AND SUGGESTIONS

5.1 Conclusion

In this work, the addition of AO25 into recycled film process dramatically decreased gel counts of recycled LLDPE film comparing with no additive added. Moreover, it can be continuously decreased until 15 phr and constant after this. In consideration of mechanical properties, e.g. tensile strength, elongation and tear resistance and yellowness index of recycled LLDPE film, it was dropped when AO25 content was increased. In addition, it was observed that higher amount of virgin grade LLDPE in recycled LLDPE film was shown to improvement of gel reduction, tensile strength, tear resistance and yellowness index. The recycled pellet of LLDPE filled with 8 phr of AO25 and virgin grade LLDPE blend at 25 percentages (P8/25) had the lowest gel counts (6 gels) and yellowness index (-7.47) and highest tensile strength (MD 2.84 and TD 2.38 kg_f), elongation (MD 911 and TD 2.38 %) and tear resistance whereas the recycled pellet LLDPE filled with 15, 25, 40 phr of AO25 and virgin grade LLDPE at 25 and 50 percentages (P15/25, P15/50, P25/25 and P40/25) were also exhibited the lowest gel counts. However, it can be concluded that P8/25 is the best composition for film due to lower AO25 loading which was the best cost effective.

5.2 Suggestions for further work

1. The addition of nitrogen as oxygen blanking should be investigated in order to monitor the effect of oxygen on gel counts in recycled LLDPE process.
2. The other antioxidant such as Irganox 1010, BHT and Vitamin E should be used and compared the gel counts between these antioxidants with AO25.
3. The influence of multi-recycled on the amount of gel counts in recycled LLDPE process should be investigated.

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APPENDICES

APPENDIX A

A. Gel counts of recycled LLDPE film.

Table A.1 Gel counts of recycled LLDPE film.

Formulation	Gel level																								Total
	0.4-0.8mm								0.8-1.5mm								>1.5mm								
	1	2	3	4	5	avg	sd	% variation	1	2	3	4	5	avg	sd	% variation	1	2	3	4	5	avg	sd	% variation	
virgin LLDPE	2	3	5	5	1	3	2	55.90	0	0	3	1	2	1	1	108.65	0	0	0	1	0	0	0	223.61	4
P0/25	10	8	5	6	7	7	2	26.72	3	5	5	4	6	5	1	24.79	0	0	0	0	0	0	0		12
P0/50	12	13	9	5	8	9	3	34.14	7	5	3	4	6	5	2	31.62	0	0	2	0	0	0	1	223.61	14
P0/75	15	12	7	8	8	10	3	33.91	7	10	11	8	6	8	2	24.69	0	0	0	0	1	0	0	223.61	18
P0/100	14	12	11	13	8	12	2	19.85	3	9	7	8	5	6	2	37.63	3	4	0	0	1	2	2	113.54	20
P3/25	1	0	3	5	2	2	2	87.43	8	7	7	6	4	6	2	23.70	0	0	0	0	0	0	0		8
P3/50	5	5	4	6	4	5	1	17.43	6	6	4	2	3	4	2	42.59	1	0	0	2	0	1	1	149.07	10
P3/75	8	9	13	15	10	11	3	26.50	0	2	3	2	2	2	1	60.86	0	2	0	0	0	0	1	223.61	13
P3/100	9	10	15	11	14	12	3	21.94	2	2	3	1	2	2	1	35.36	1	1	1	2	1	1	0	37.27	15
P8/25	2	3	4	6	5	4	2	39.53	0	0	3	4	2	2	2	99.38	0	1	1	0	0	0	1	136.93	6
P8/50	11	11	6	7	6	8	3	31.57	1	4	2	1	3	2	1	59.27	0	0	0	0	0	0	0		10
P8/75	6	8	7	8	6	7	1	14.29	5	7	2	4	3	4	2	45.80	0	1	2	2	1	1	1	69.72	12
P8/100	7	6	10	8	8	8	1	19.02	0	4	1	3	2	2	2	79.06	2	2	3	1	4	2	1	47.51	12
P15/25	3	4	6	2	4	4	1	39.03	3	2	1	3	3	2	1	37.27	0	0	0	1	0	0	0	223.61	6
P15/50	3	4	4	2	1	3	1	46.57	3	3	4	2	4	3	1	26.15	0	0	1	0	1	0	1	136.93	6
P15/75	5	8	8	8	9	8	2	19.95	0	3	0	1	2	1	1	108.65	0	0	0	0	0	0	0		9
P15/100	7	10	10	9	6	8	2	21.63	0	0	1	0	0	0	0	223.61	1	1	2	1	0	1	1	70.71	9
P25/25	1	3	3	0	4	2	2	74.69	6	6	2	4	3	4	2	42.59	0	1	0	0	1	0	1	136.93	6
P25/50	2	2	3	1	3	2	1	38.03	3	5	4	6	7	5	2	31.62	0	0	0	0	0	0	0		7
P25/75	5	6	5	8	10	7	2	31.88	2	1	2	3	3	2	1	38.03	0	2	0	0	0	0	1	223.61	9
P25/100	2	6	4	4	5	4	1	35.32	8	3	5	4	6	5	2	36.99	0	0	0	0	0	0	0		9
P40/25	2	1	5	3	3	3	1	52.97	0	4	2	2	1	2	1	82.40	0	1	3	1	1	1	1	91.29	6
P40/50	3	3	4	5	2	3	1	33.53	4	3	5	4	6	4	1	25.91	0	0	0	2	0	0	1	223.61	7
P40/75	3	3	2	2	4	3	1	29.88	6	6	6	4	5	5	1	16.56	0	0	0	0	0	0	0		8
P40/100	4	4	5	2	5	4	1	30.62	1	3	3	5	2	3	1	52.97	1	3	1	0	1	1	1	91.29	8
result 4.1																									
175 °C	32	48	55	52	38	45	10	21.55	10	9	4	7	6	7	2	33.16	2	0	2	3	1	2	1	71.26	54
190 °C	15	19	24	27	17	20	5	24.41	5	4	5	6	8	6	2	27.08	0	0	2	1	0	1	1	149.07	27
210 °C	12	13	18	22	24	18	5	29.83	9	8	11	14	8	10	3	25.50	2	4	3	2	5	3	1	40.75	31
230 °C	24	38	29	33	37	32	6	18.03	13	11	7	9	7	9	3	27.74	0	2	3	1	0	1	1	108.65	42
250 °C	30	36	37	24	26	31	6	19.00	10	11	11	13	7	10	2	21.07	3	4	2	5	2	3	1	40.75	44
result 4.2																									
waste LLDPE	7	6	2	3	3	4	2	51.62	0	0	1	1	0	0	1	136.93	0	0	0	2	0	0	1	223.61	4
recycled LLDPE	12	8	6	11	7	9	3	29.41	3	4	1	2	4	3	1	46.57	2	2	1	5	1	2	2	74.69	14

APPENDIX B

B. Tensile properties of recycled LLDPE film

Table B.1 Tensile strength at break (kgf/mm^2)

Sample No.	SIDE	Tensile(Kg$_f$/mm^2)							
		1	2	3	4	5	AVG	SD	%variation
1210 100%	MD	2.72	2.90	3.20	3.15	3.06	3.01	0.19	6.47
	TD	2.89	3.07	2.90	3.07	3.03	2.99	0.09	2.96
P0/25	MD	3.01	2.64	2.88	3.13	2.86	2.90	0.19	6.40
	TD	2.57	2.29	2.22	2.48	2.41	2.39	0.14	5.95
P0/50	MD	2.60	2.29	2.67	2.64	2.55	2.55	0.15	5.91
	TD	2.25	2.28	2.09	2.06	2.07	2.15	0.10	4.88
P0/75	MD	2.50	2.18	2.16	2.18	2.24	2.25	0.14	6.22
	TD	1.98	1.83	2.15	2.26	1.81	2.01	0.20	9.92
P0/100	MD	2.25	2.36	2.05	2.22	2.03	2.18	0.14	6.41
	TD	1.96	2.03	1.92	1.95	2.08	1.99	0.07	3.33
P3/25	MD	2.96	3.01	2.65	3.01	3.05	2.94	0.16	5.61
	TD	2.75	2.70	2.47	2.42	2.37	2.54	0.17	6.73
P3/50	MD	2.68	2.89	2.57	2.56	2.69	2.68	0.13	4.96
	TD	2.18	2.14	2.08	2.34	2.50	2.25	0.17	7.59
P3/75	MD	2.18	2.29	2.20	2.49	2.29	2.29	0.12	5.24
	TD	2.12	2.18	2.04	1.87	2.13	2.07	0.12	5.91
P3/100	MD	2.28	2.06	2.08	2.32	2.41	2.23	0.15	6.88
	TD	2.06	2.02	2.07	1.95	1.88	1.99	0.08	4.08
P8/25	MD	2.58	3.13	2.78	2.78	2.91	2.84	0.20	7.15
	TD	2.70	2.06	2.31	2.47	2.37	2.38	0.23	9.67
P8/50	MD	2.38	2.54	2.30	2.42	2.55	2.44	0.11	4.45
	TD	1.97	1.89	2.01	2.28	2.29	2.09	0.19	8.94
P8/75	MD	2.17	2.30	2.11	2.00	2.32	2.18	0.14	6.20
	TD	2.07	2.02	1.80	1.96	2.06	1.98	0.11	5.45
P8/100	MD	2.03	2.24	1.90	1.90	2.13	2.04	0.15	7.28
	TD	2.03	1.79	1.66	1.82	1.88	1.83	0.13	7.28
P15/25	MD	2.38	2.42	2.07	2.03	1.87	2.15	0.24	10.96
	TD	1.66	2.05	2.01	2.02	2.14	1.98	0.19	9.37
P15/50	MD	2.12	1.60	2.05	2.40	2.33	2.10	0.32	15.06
	TD	1.71	1.73	2.00	2.16	2.01	1.92	0.20	10.22
P15/75	MD	1.73	2.09	2.07	2.19	2.12	2.04	0.18	8.90
	TD	1.80	1.92	1.75	1.89	1.90	1.85	0.07	3.89
P15/100	MD	1.92	1.88	1.89	1.88	1.70	1.85	0.09	4.69
	TD	2.13	1.78	1.83	1.96	1.81	1.90	0.14	7.57
P25/25	MD	2.18	2.08	1.83	2.20	2.37	2.13	0.20	9.27
	TD	1.98	2.01	2.05	1.76	1.88	1.93	0.12	6.00
P25/50	MD	2.24	2.08	1.80	2.00	1.92	2.01	0.17	8.31
	TD	1.64	1.86	1.89	1.96	1.62	1.79	0.16	8.68
P25/75	MD	1.82	1.83	2.10	2.17	2.00	1.98	0.16	7.96
	TD	1.68	1.44	1.70	1.80	1.80	1.68	0.15	8.72
P25/100	MD	2.42	2.03	1.68	1.76	2.06	1.99	0.29	14.68
	TD	1.60	1.95	1.65	1.62	1.84	1.73	0.16	8.96

Table B.1 Tensile strength at break (kgf/mm^2)(continued)

Sample No.	SIDE	Tensile(Kg$_f$/mm^2)							
		1	2	3	4	5	AVG	SD	%variation
P40/25	MD	1.66	1.94	1.80	1.90	1.80	1.82	0.11	5.99
	TD	1.71	1.49	1.83	1.50	1.76	1.66	0.15	9.29
P40/50	MD	1.55	1.76	1.65	1.57	1.47	1.60	0.11	6.91
	TD	1.60	1.33	1.68	1.73	1.69	1.61	0.16	10.05
P40/75	MD	1.65	1.63	1.80	1.72	1.39	1.64	0.15	9.22
	TD	1.38	1.59	1.58	1.81	1.79	1.63	0.18	10.86
P40/100	MD	1.88	1.69	1.33	1.34	1.42	1.53	0.24	15.89
	TD	1.23	1.37	1.21	1.34	1.31	1.29	0.07	5.28
Result of4.1									
175^{o}C	MD	2.92	3.00	2.99	3.18	3.12	3.04	0.10	3.45
	TD	2.71	2.64	2.78	2.62	2.88	2.73	0.11	3.91
190^{o}C	MD	3.39	3.23	3.35	3.39	3.47	3.37	0.08	2.51
	TD	3.00	2.83	2.84	2.90	2.95	2.90	0.07	2.48
210^{o}C	MD	3.07	3.43	3.07	3.53	3.27	3.27	0.21	6.43
	TD	3.07	3.11	2.86	2.88	2.81	2.95	0.13	4.58
230^{o}C	MD	3.05	3.21	2.97	3.28	3.09	3.12	0.12	3.99
	TD	2.68	2.68	2.85	3.03	2.69	2.78	0.15	5.54
250^{o}C	MD	2.94	2.74	2.90	2.63	2.46	2.73	0.20	7.26
	TD	2.21	2.36	2.46	2.20	2.27	2.30	0.11	4.70
Result of4.3									
waste LLDPE	MD	2.34	2.51	2.18	2.17	2.21	2.28	0.15	6.41
	TD	1.89	2.04	2.14	2.26	2.40	2.15	0.20	9.15
Recycle LLDPE	MD	1.95	2.07	2.38	2.22	2.29	2.18	0.17	7.92
	TD	1.67	2.08	2.04	2.02	2.04	1.97	0.17	8.62

Table B.2 Elongation at break (%)

Sample No.	SIDE	Elongation at break (%)							
		1	2	3	4	5	AVG	SD	%variation
1210 100%	MD	1071	1037	1062	1033	1052	1051	16.14	1.54
	TD	1125	1058	992	1079	1033	1057	49.77	4.71
P0/25	MD	808.4	879.2	1033	966.7	995.8	937	91.42	9.76
	TD	987.3	966.7	1017	1079	1087	1027	53.89	5.25
P0/50	MD	933.4	883.4	887.5	962.7	879.2	909	37.03	4.07
	TD	996.1	1017	966.7	1034	1029	1009	27.59	2.74
P0/75	MD	900.1	841.9	791.6	829.6	883.5	849	43.37	5.11
	TD	978.9	858.4	970.6	1012	928.9	950	59.03	6.22
P0/100	MD	871.3	729.7	708.9	798.3	833.7	788	68.54	8.69
	TD	995.4	1020	941.3	816.8	887.2	932	82.35	8.83
P3/25	MD	945.9	987.4	900	904.2	946	937	35.86	3.83
	TD	1137	1112	1017	1013	1000	1056	63.65	6.03
P3/50	MD	933.4	883.4	887.5	962.7	879.2	909	37.03	4.07
	TD	933.8	958.3	1033	1050	1088	1013	64.53	6.37
P3/75	MD	1016	879	800	783	783.1	852	99.80	11.71
	TD	975.2	966.7	916.9	942	975.3	955	25.38	2.66
P3/100	MD	779.1	796.1	783.6	804.7	804.4	794	11.79	1.49
	TD	941.5	925.2	995.5	999.7	870.9	947	53.44	5.65
P8/25	MD	879.3	958.3	900	887.6	929.1	911	32.55	3.57
	TD	1054	1017	1041	999.8	958.6	1014	37.45	3.69
P8/50	MD	920.6	829.3	912.5	895.9	879.3	888	36.21	4.08
	TD	1065	896.1	954.3	979.4	958.5	971	61.14	6.30
P8/75	MD	766.7	817	758.1	820.9	745.8	782	34.83	4.46
	TD	974.8	912.2	948.3	866.5	937.4	928	41.00	4.42
P8/100	MD	762.5	766.6	808.7	746.1	695.8	756	40.79	5.40
	TD	779	833.1	887.3	912.1	900	862	55.49	6.44
P15/25	MD	816.9	762.8	804.2	792.3	708.8	777	43.07	5.54
	TD	835.8	945.9	916.9	945.9	953.9	920	48.96	5.32
P15/50	MD	662.2	787.4	758.2	783.2	799.8	758	55.73	7.35
	TD	887.1	758.2	899.7	949.6	874.6	874	70.64	8.08
P15/75	MD	762.6	737.9	679	770.7	799.9	750	45.46	6.06
	TD	820.6	870.6	824.6	774.8	874.7	833	41.12	4.94
P15/100	MD	738	725	771	775	742	750	21.79	2.90
	TD	822	850	843	822	805	828	18.09	2.18
P25/25	MD	812.6	782.8	721.2	716.8	766.7	760	40.94	5.39
	TD	913.3	870	884	920	887	895	21.03	2.35
P25/50	MD	792.7	829.5	746.2	696.2	673.2	748	65.04	8.70
	TD	746.3	762.8	841.4	895.6	878.6	825	67.43	8.17
P25/75	MD	754.1	742.1	729	716.9	762.6	741	18.47	2.49
	TD	733.1	874.7	758.3	864.8	787.6	804	63.40	7.89
P25/100	MD	799.8	712.4	721	716.7	762.3	742	37.78	5.09
	TD	704.2	862.4	804	854.7	895.7	824	74.68	9.06
P40/25	MD	737.4	729.1	703.9	699.9	799.9	734	40.15	5.47
	TD	766.5	816.4	820.7	874.7	724.8	801	57.12	7.14
P40/50	MD	650.1	554	662.7	724.7	729.1	664	71.09	10.70
	TD	792.1	800.4	800.4	738	792.2	785	26.39	3.36
P40/75	MD	675.3	712.5	670.7	670.8	654.6	677	21.46	3.17
	TD	887.7	679.2	833.1	783.6	808.4	798	76.98	9.64
P40/100	MD	587.4	624.9	624.9	645.6	595.8	616	23.77	3.86
	TD	733.5	691.6	795.5	716.4	779	743	43.28	5.82

Table B.2 Elongation at break (%) (continued)

Sample No.	SIDE	Elongation at break (%)							
		1	2	3	4	5	AVG	SD	%variation
Result of4.1									
175°C	MD	904.4	933.6	925.2	971.1	996.3	946	37.00	3.91
	TD	1133	1138	1158	1142	1287	1172	65.19	5.56
190°C	MD	1108	1088	1050	1004	1108	1072	44.60	4.16
	TD	1204	1154	1196	1187	1196	1187	19.62	1.65
210°C	MD	1038	1175	1196	1179	1154	1148	63.50	5.53
	TD	1229	1288	1141	1220	1158	1207	59.10	4.90
230°C	MD	1158	954.1	1100	1062	1088	1072	74.89	6.98
	TD	1096	1071	1075	1133	1058	1087	29.31	2.70
250°C	MD	900	879.6	883.7	871.1	808.6	869	35.15	4.05
	TD	966.9	975.1	1013	954.4	975.3	977	21.88	2.24
Result of4.3									
waste LLDPE	MD	702.7	768.1	749	804.3	713	747	41.38	5.54
	TD	986.2	994.7	945.2	921.3	904.5	950	39.45	4.15
Recycle LLDPE	MD	621.7	674	695.4	688.4	700	676	31.85	4.71
	TD	814.8	887.7	791.3	807.3	817.4	824	37.19	4.52

APPENDIX C

C. Tear resistance of recycled LLDPE film

Table C.1 Tear resistance of recycled LLDPE film

Sample No.	SIDE	Tear resistance (kgf)							
		1	2	3	4	5	AVG	SD	%variation
1210 100%	MD	0.92	0.88	0.93	0.86	0.90	0.90	0.03	3.48
	TD	0.92	0.84	0.84	0.92	0.91	0.89	0.05	5.08
P0/25	MD	0.85	0.79	0.83	0.84	0.84	0.83	0.03	3.12
	TD	0.85	0.86	0.85	0.91	0.93	0.88	0.04	4.03
P0/50	MD	0.80	0.83	0.82	0.75	0.81	0.80	0.03	4.13
	TD	0.89	0.79	0.82	0.86	0.91	0.85	0.05	6.20
P0/75	MD	0.79	0.78	0.81	0.76	0.77	0.78	0.02	2.42
	TD	0.84	0.87	0.84	0.83	0.86	0.85	0.02	1.99
P0/100	MD	0.81	0.80	0.76	0.76	0.77	0.78	0.03	3.30
	TD	0.83	0.81	0.84	0.82	0.80	0.82	0.02	2.04
P3/25	MD	0.83	0.88	0.86	0.79	0.81	0.83	0.04	4.47
	MD	0.87	0.88	0.89	0.89	0.89	0.88	0.01	0.91
P3/50	MD	0.74	0.80	0.80	0.81	0.88	0.81	0.05	6.22
	TD	0.91	0.86	0.91	0.81	0.84	0.87	0.04	4.96
P3/75	MD	0.78	0.78	0.86	0.73	0.82	0.79	0.05	6.22
	TD	0.85	0.83	0.81	0.87	0.85	0.84	0.02	2.60
P3/100	MD	0.74	0.81	0.79	0.80	0.77	0.78	0.03	3.64
	TD	0.83	0.82	0.82	0.82	0.85	0.83	0.01	1.61
P8/25	MD	0.92	0.74	0.83	0.83	0.79	0.82	0.06	7.78
	TD	0.86	0.88	0.88	0.86	0.88	0.87	0.01	1.35
P8/50	MD	0.70	0.83	0.76	0.81	0.87	0.80	0.06	8.15
	TD	0.93	0.78	0.77	0.78	0.99	0.85	0.10	12.08
P8/75	MD	0.78	0.78	0.77	0.75	0.82	0.78	0.02	3.11
	TD	0.84	0.83	0.83	0.82	0.80	0.82	0.02	1.97
P8/100	MD	0.73	0.75	0.74	0.77	0.73	0.74	0.02	2.24
	TD	0.81	0.81	0.85	0.77	0.76	0.80	0.04	4.62
P15/25	MD	0.73	0.72	0.84	0.79	0.75	0.76	0.05	6.22
	TD	0.81	0.82	0.77	0.80	0.86	0.81	0.03	4.03
P15/50	MD	0.72	0.76	0.72	0.74	0.84	0.75	0.05	6.58
	TD	0.77	0.78	0.79	0.86	0.81	0.80	0.03	4.32
P15/75	MD	0.74	0.70	0.71	0.70	0.75	0.72	0.02	3.14
	TD	0.78	0.79	0.79	0.79	0.79	0.79	0.01	0.64
P15/100	MD	0.70	0.70	0.72	0.73	0.69	0.71	0.02	2.58
	TD	0.77	0.74	0.77	0.78	0.75	0.76	0.01	1.74
P25/25	MD	0.78	0.75	0.74	0.76	0.75	0.76	0.02	2.22
	TD	0.72	0.86	0.81	0.76	0.86	0.80	0.06	7.94
P25/50	MD	0.72	0.67	0.72	0.73	0.71	0.71	0.02	3.23
	TD	0.78	0.81	0.79	0.75	0.79	0.78	0.02	2.74
P25/75	MD	0.66	0.78	0.68	0.68	0.68	0.70	0.05	6.74
	TD	0.78	0.76	0.75	0.79	0.76	0.77	0.02	2.00
P25/100	MD	0.68	0.71	0.72	0.72	0.68	0.70	0.02	3.01
	TD	0.77	0.76	0.78	0.80	0.78	0.78	0.02	2.03

Table C.1 Tear resistance of recycled LLDPE film (continued)

Sample No.	SIDE	Tear resistance (kgf)							
		1	2	3	4	5	AVG	SD	%variation
P40/25	MD	0.67	0.70	0.74	0.67	0.64	0.69	0.04	5.70
	TD	0.70	0.70	0.68	0.72	0.70	0.70	0.01	1.57
P40/50	MD	0.62	0.61	0.64	0.60	0.64	0.62	0.02	2.69
	TD	0.66	0.67	0.63	0.63	0.72	0.66	0.04	5.36
P40/75	MD	0.64	0.64	0.62	0.64	0.62	0.63	0.01	1.34
	TD	0.68	0.68	0.67	0.70	0.72	0.69	0.02	2.57
P40/100	MD	0.59	0.59	0.57	0.62	0.60	0.59	0.02	3.37
	TD	0.64	0.63	0.63	0.64	0.61	0.63	0.01	1.94
Result of4.3									
waste LLDPE	MD	0.85	0.81	0.75	0.89	0.76	0.81	0.06	7.08
	TD	0.71	0.75	0.77	0.75	0.75	0.75	0.02	3.02
Recycle LLDPE	MD	0.62	0.74	0.71	0.70	0.64	0.68	0.05	7.48
	TD	0.63	0.66	0.65	0.70	0.65	0.66	0.03	4.12

APPENDIX D

D. Yellowness index of recycled LLDPE film

Table D.1 Yellowness index of recycled LLDPE film

Sample No.	Yellowness Index							
	1	2	3	4	5	AVG	SD	%variation
1210 100%	-7.59	-7.81	-7.64	-7.81	-7.73	-7.72	0.10	-1.29
P0/25	-7.68	-7.34	-7.45	-7.59	-7.62	-7.54	0.14	-1.84
P0/50	-7.42	-7.34	-7.45	-7.59	-7.62	-7.48	0.12	-1.58
P0/75	-7.21	-7.42	-7.28	-7.44	-7.1	-7.29	0.14	-1.96
P0/100	-7.31	-7.14	-7.3	-7.12	-7.33	-7.24	0.10	-1.40
P3/25	-7.75	-7.52	-7.34	-7.55	-7.74	-7.58	0.17	-2.25
P3/50	-7.44	-7.36	-7.58	-7.05	-7.32	-7.35	0.19	-2.65
P3/75	-7.18	-7.28	-7.08	-7.23	-7.33	-7.22	0.10	-1.33
P3/100	-6.9	-7.01	-7	-6.94	-6.97	-6.96	0.05	-0.65
P8/25	-7.33	-7.41	-7.56	-7.61	-7.44	-7.47	0.11	-1.52
P8/50	-6.89	-6.85	-6.75	-6.97	-7.03	-6.90	0.11	-1.57
P8/75	-6.63	-6.82	-6.58	-6.77	-6.71	-6.70	0.10	-1.47
P8/100	-6.55	-6.43	-6.36	-6.32	-6.45	-6.42	0.09	-1.38
P15/25	-7.02	-6.98	-6.84	-7.12	-7.06	-7.00	0.11	-1.50
P15/50	-6.751	-6.95	-6.82	-6.74	-7.01	-6.85	0.12	-1.76
P15/75	-6.62	-6.35	-6.59	-6.72	-6.84	-6.62	0.18	-2.74
P15/100	-6.39	-6.45	-6.46	-6.46	-6.3	-6.41	0.07	-1.08
P25/25	-6.82	-6.72	-6.98	-6.55	-6.91	-6.80	0.17	-2.48
P25/50	-6.18	-6.35	-6.02	-6.28	-6.23	-6.21	0.12	-2.00
P25/75	-6.03	-6.12	-6.27	-6.89	-6.87	-6.44	0.41	-6.44
P25/100	-5.81	-5.91	-5.93	-5.8	-5.9	-5.87	0.06	-1.03
P40/25	-6.82	-6.52	-6.63	-6.42	-6.62	-6.60	0.15	-2.25
P40/50	-6.13	-6.28	-6.35	-6.43	-6.24	-6.29	0.11	-1.80
P40/75	-5.87	-6.01	-6.24	-6.03	-5.94	-6.02	0.14	-2.31
P40/100	-5.83	-5.87	-5.81	-5.86	-5.74	-5.82	0.05	-0.89
Result of4.3								
waste LLDPE	-9.01	-8.95	-9.12	-9.32	-8.96	-9.07	0.15	-1.70
Recycle LLDPE	-6.85	-7.12	-7.05	-7.07	-7.21	-7.06	0.13	-1.88

APPENDIX E

E. Melt flow index of recycled LLDPE film

Table E.1 Melt flow index of recycled LLDPE film

Sample No.	Melt Flow Index (g/10 min)							
	1	2	3	4	5	AVG	SD	%variation
P0	0.95	0.98	1	1.19	0.99	1.02	0.10	9.37
P3	1.1	1.12	1.01	0.98	1.2	1.08	0.09	8.17
P8	1.12	1.31	1.22	1.27	1.23	1.23	0.07	5.78
P15	1.71	1.42	1.52	1.61	1.71	1.59	0.13	7.87
P25	1.74	1.82	1.91	1.66	1.72	1.77	0.10	5.48
P40	2.21	2.25	2.08	2.03	2.08	2.13	0.09	4.44

VITAE

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