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**COMPATIBILIZED POLY(ETHYLENE TEREPHTHALATE)/
EPDM BLENDS**



Miss Pornrut Rungraungthitisuk

**สถาบันวิทยบริการ
จุฬาลงกรณ์มหาวิทยาลัย**
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การศึกษาพอลิเมอร์ผสมพอลิเอทิลีนเทเรฟทาเลตและอีพีดีเอ็มโดยใช้อีพีเอ็มกราฟต์มาลิกแอนไฮโดรด์เป็นสารประสาน โดยศึกษาผลของปริมาณมาลิกแอนไฮโดรด์ อัตราส่วนการผสม ปริมาณไดอินและความหนืดมูนนี้ของอีพีดีเอ็มต่อสมบัติพอลิเมอร์ผสม อีพีดีเอ็มกราฟต์มาลิกแอนไฮโดรด์ที่เตรียมได้ผสมกับพอลิเอทิลีนเทเรฟทาเลตโดยใช้เครื่องผสมสกรูแบบตัวหนอนเดี่ยว ที่อุณหภูมิ 250 องศาเซลเซียส และความเร็วรอบ 50 รอบต่อนาที พอลิเมอร์ผสมพอลิเอทิลีนเทเรฟทาเลตและอีพีดีเอ็มที่ไม่มีมาลิกแอนไฮโดรด์ให้สมบัติเชิงกลต่ำเนื่องจากมีแรงกระทำระหว่างเฟสของพอลิเมอร์และเฟสของอีลาสโตเมอร์ต่ำ การเติมอีพีดีเอ็มกราฟต์มาลิกแอนไฮโดรด์ในพอลิเมอร์ผสมเพิ่มความเข้ากันได้ของพอลิเมอร์ผสมพอลิเอทิลีนเทเรฟทาเลตและอีพีดีเอ็มโดยเกิดแรงกระทำและพันธะเคมีที่มากขึ้นระหว่างเฟสของพอลิเมอร์ผสม ทำให้ความสามารถในการไหลของพอลิเมอร์เหลวของพอลิเมอร์ผสมลดลงและสมบัติการทนแรงกระทำเพิ่มขึ้น สมบัติการทนต่อแรงดึงและการยืดตัวเมื่อขาดของพอลิเมอร์ผสมพอลิเอทิลีนเทเรฟทาเลตและอีพีดีเอ็มที่มีอีพีดีเอ็มกราฟต์มาลิกแอนไฮโดรด์มีค่าต่ำลงเมื่อเทียบกับพอลิเอทิลีนเทเรฟทาเลต โครงสร้างเฟสของพอลิเมอร์ผสมแสดงหยดของอีพีดีเอ็มที่กระจายตัวอยู่ในพอลิเอทิลีนเทเรฟทาเลตเมทริกซ์ สำหรับพอลิเมอร์ผสมที่มีอีพีดีเอ็มกราฟต์มาลิกแอนไฮโดรด์เป็นสารประสาน หยดของอีพีดีเอ็มมีการกระจายตัวดีขึ้นและขนาดเล็กลงเมื่อเพิ่มปริมาณของมาลิกแอนไฮโดรด์

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The blending of PET and EPDM using EPDM-g-MAH as compatibilizer was studied. The effect of MAH contents, blending ratio, diene content and Mooney viscosity of EPDM on the blend properties was investigated. Grafting of MAH onto EPDM was prepared and then EPDM-g-MAH was blended with PET by single screw extruder at 250°C and 50 rpm. PET/EPDM blend without grafted MAH produced poor mechanical properties because the interaction between polymer phase and elastomer phase was weak. The using of EPDM-g-MAH in the blend increased the compatibility of PET-EPDM system by generating higher interaction and chemical link between interfacial surface of the blends. Consequently, the melt flow index of polymer blends decreased and the impact strength increased. The tensile strength and elongation at break of PET/EPDM blend with EPDM-g-MAH was lower than pure PET. The morphology of the blends showed the droplets of EPDM dispersed in PET matrix. For the blend with EPDM-g-MAH compatibilizer, the droplets were finer and smaller with increasing MAH content.

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CHAPTER I

INTRODUCTION

1.1 Introduction

Poly(ethylene terephthalate); PET is unsaturated polyester of terephthalic acid and ethylene glycol. According to a recent forecast, the consumption of poly(ethylene terephthalate) will rise by 10% per year in the near future [1]. The major reason for this optimistic estimate is the replacement of glass and aluminum containers by PET-based ones, the advantages of PET bottles arise from their light weight and resistance to shatter, accompanied with further benefits in transportation, distribution and other related costs. Returnable and one-way PET bottles are widely used at present for the packaging of the soft drinks, vegetable oils and even of beer. The majority of discarded PET bottles became part of the municipal waste solid stream destined for disposal via landfill and incineration, both of which are environmentally unfriendly practices, because of the irrecoverable loss of non-renewable energy-rich resource. Recycling aim at the use of plastic parts in high value applications requires that properties of the recycled material are tailor-made for targeted product and end-use. They enable the manufacture to produce suitable materials for application in e.g., the electric, automotive, building, and consumer goods areas. The development of appropriate formulations and techniques for the mechanical recycling of post consumer waste in high-end application. Mechanical recycling of post consumer plastic waste make economical, ecological and social political sense only if an efficient system of collecting and sorting the waste has been well conceived and implement. Although 'closed loop' recycling (i.e. bottle from bottle) is an ideal operation for recovering value from discarded PET bottles, its use has been limited because of PET is undergo thermal and hygrothermal degradation and the stringent safety requirements placed upon plastics destined for food contact applications.

PET is sensitive to chain scission, whether induced or catalyzed by heat, water, acidic and caustic components present as labels, label glues, or used for bottles cleaning prior to refill, its molecular weight is inevitably reduced during service and

the major task to be solved with respect to recycling is to increase, to restore molecular weight, or develop to PET-based blends and alloys. Melt blending of PET with various polymers and elastomer is offered a good opportunity to convert the recycled PET into high performance thermoplastics.

The overall aim of this study is to convert post-consumer PET bottles into value-added thermoplastic blend with ethylene propylene copolymer (EPDM) and ethylene propylene copolymer grafted maleic anhydride (EPDM-g-MAH) as a compatibilizer and produce a new material with an upgraded property.

1.2 Objective

To study the properties of blend of poly(ethylene terephthalate) and ethylene propylene copolymer by using ethylene propylene copolymer grafted maleic anhydride (EPDM-g-MAH) as compatibilizer.

1.3 Scope of the Research

1. Literature survey and in-depth study of the research work.
2. Preparation of EPDM (Buna6470)-g-MAH using laboratory single screw extruder. Effect of maleic anhydride contents (0, 1, 3, and 5 phr) on the percentage of maleic anhydride grafted onto EDPM was studied.
3. Preparation of EPDM(Buna 3590)-g-MAH using laboratory single screw extruder. Effect of maleic anhydride contents (0, 1, 3, and 5 phr) on the percentage of maleic anhydride grafted onto EDPM was studied.
4. Blends of PET/EPDM with and without EPDM-g-MAH were prepared. The effects of blend compositions (80/20 and 70/30) were studied.
5. Determination of tensile strength and impact strength of PET/EPDM blend and evaluation of the blend miscibility and the morphology using the techniques of differential scanning calorimetry (DSC) and scanning electron microscopy (SEM).
6. Summarizing the results.

CHAPTER II

THEORY AND LITERATURE REVIEW

Recycling operations, which have proliferated in recent years, are providing an abundant and inexpensive source for some thermoplastics. For instance, the recycling in the U.S. for plastic soft drink bottles made out of polyethylene terephthalate topped 31% in 1990 [2]. Recycled polyethylene terephthalate is particularly useful as an injection moldable material which can be formed into articles exhibiting a good balance of properties, including strength and stiffness. However, an improvement in impact strength and processability of these recycled PET materials is desirable. Use of recycled PET in food contact application is supported by a range of factor including recycled content legislation in some countries, the drive to create recycling, high value applications for recycled PET.

The methods for improvement of recycled PET are using chain extender compound that are able to react with the functional group of PET in melt. The reactivity of chain extender should be based on functional end group. Considering the type of the functional groups in the chain extenders listed above, one can easily outline a straightforward strategy for the reactive compatibilization of PET/ polyolefin blends, the compatibilizers should have the same functional groups (anhydride, epoxy, oxazoline, etc) as the chain extender [1]. Another method, the commercialization of PET will fuel the future development of PET-based blends and alloys for manufacturing. The last method for developing PET is blending with rubber to conventional thermoplastic elastomers.

However, physical blending of these polymers have not been successful. Since polyolefins and thermoplastic polyesters are naturally incompatible, mere mixing of these polymers involves problems. These immiscible plastics exhibit poor adhesion along blend interface, with result of weakness. The mechanical properties, in

particular, impact resistance, tensile elongation, and tensile strength of a mold product made of mixture of polyblends often have values lower than these expected by simple additivity or averaging of the physical properties of the polyolefin and thermoplastic polyester. The processibility of the mixture of polymer blend is limited. So interfacial agents are required to produce multipolymer blends with a desirable balance of properties. Interfacial agent provides adhesion between the principal phase, improving stress transfer, and are necessary to reduce interfacial tension during processing that can lead to gross phase separation. Thus, interfacial agents play an important role in determining the ultimate morphology of the blend.

2.1. Polymer Blending

Polymer blending are two polymer mix together in order to get a material with properties somewhere between those of two polymers mixed. Different polymer blends when together may be miscible, partially miscible, or completely immiscible. The methods of blending may be polymerization blending, suspension blending, fines power mixing and melt blending [3]. Polymer melt blending can offer a convenient way to develop new materials easily in which the properties of the blend components are combined, are non-toxic environment. So almost polymer blending is carried out by mechanical mixing of molten polymers. Disentanglement and viscous flow are difficult, so temperature, shear, and time are major factors, and final quenching may freeze-in nonequilibrium morphology. Generally, increasing temperature may increase or decrease thermodynamic miscibility. It will always lower viscosity, though the quantitative effects on the two phases may be different and it will certainly accelerate the approach to equilibrium; but conversely, return to room temperature may produce a serious shift in equilibrium. In the same way, increasing shear will decrease domain size, within the limits permitted by melt viscosity. Directional shear vectors can convert spherical domains in to fibrillar morphology, which might contribute mechanical reinforcement to matrix polymer, or into lamellar morphology, which has been used to reduce permeability; and of course the same vectors can produce orientation, which would enhance these effects. Extreme shear forces, particularly at low temperature and very high viscosities, can actually break otherwise stable polymer molecules into macroradicals. When A-A and B-B polymer molecules are sheared into macroradicals $A\cdot$ and $B\cdot$, cross-combination of these radicals can

then produce A-B block or graft copolymer, which would properly be classified later as reactive compatibilization.

When either polymer crystallizes during cooling of the melt, the crystals cause of a separate phase. When both polymers crystallize, this can add two phases to the system. In rare cases, when both polymers have similar crystal structure, they may be able to cocrystallize; when this happens, is actually contributed to intermolecular attraction and compatibilization [4].

In the past few decades, many polymers have been found to be miscible with dissimilar polymers, although immiscibility of pair of high molecular weight polymer was predicted, except for the pair possessing polar substituents that interact favorably with one another. In order to achieve compatibilization of polymer blends, whether by addition of a third component or by inducing *in situ* chemical reaction between blend components (reactive blending), has an important role in the development of polymer blends [5].

2.2. Miscibility and Compatibility

Compatibility is frequently defined as miscibility on a molecular scale. Another way of definitions compatible blends is polymer mixtures, which are not exhibited gross symptoms of phase separation; most compatibilized blends contain very finely dispersed phase. A third definition preferred by passes a desirable set of properties. The achievement of compatibilization, whether by addition of a third component (a so-called 'compatibilizer) or by inducing *in situ* chemical reaction between blends components (reactive blending), has played an important role in the development of polymer blends.

2.2.1 Compatibilization Mechanisms

In most case, melt mixing two polymers results in blends which are weak and brittle; while the low deformation modulus may follow an approximately linear mixing rule, the ultimate properties certainly will not. This is because the incorporation of dispersed phase in a matrix leads to the presence of stress contents and weak interface, arising from poor mechanical coupling between phase. The factor contributing to end use properties during manufacture of a blend by melt

compounding, and subsequent conversion processing to produce a finished article, are illustrated in Figure 2.1.

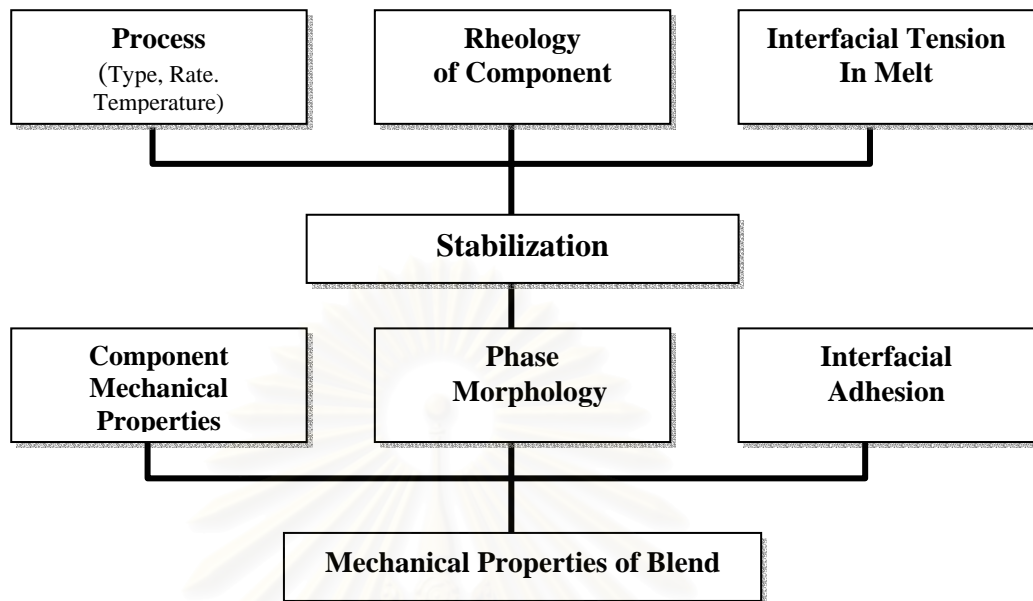


Figure 2.1 Summary of the factor contributing to end-use properties in melt [3].

The mechanical properties of a blend will be determined not only by the properties of its components, but also by the phase morphology and the interphase adhesion, both of which are important from the viewpoint of stress transfer within the blend in its end-use application. The phase morphology will normally be determined by factors as process variables (mixer type, rate of mixing, and temperature history), the rheology of the blend components and interfacial tension between phase in the melt are important. The phase morphology is unlikely to be in thermodynamic equilibrium, but generally will have been stabilized against de-mixing by some methods; this usually means via quenching to below glass transition temperature of one or both phases, or via occurrence of crystallinity in one or both phases, or occasionally by crosslinking.

The compatibilization can in principle interact in complex ways to influence final blend properties. One effect of compatibilizers is to reduce the interfacial tension in the melt, causing an emulsifying effect and leading to extremely fine dispersion of one phase in another. Another effect is to increase the adhesion at phase boundaries,

giving improved stress transfer. A third effect is to stabilize the dispersed phase against growth during annealing, again by modifying the phase –boundary interface.

2.2.2 Methods of Compatibilization

Technological definition of compatibilization as modification of blends to produce a desirable set of properties, a number of different lines of approach can be defined which may assist the materials developer. Broadly, these are:

- a) Achievement of thermodynamic miscibility.
- b) Addition of block and graft copolymers.
- c) Addition of functional/reactive polymers.
- d) In situ grafting/polymerization (reactive blending).

a) Achievement of Thermodynamic Miscibility

Compatibilization by the achievement of thermodynamic miscibility is a concept, which has been exploited in only a handful of situation to produce commercial blends. The miscibility between polymers is determined by a balance of enthalpic and entropic contributions to the free energy of mixing. While for small molecules the entropy is high enough to ensure miscibility, for polymer the entropy is almost zero, causing entropy to be decisive in determining miscibility. The change in free energy on mixing (ΔG) is writing as

$$\Delta G = \Delta H - T\Delta S \quad (2.1)$$

where H is enthalpy, S is entropy and T is temperature. For spontaneous mixture, ΔG must be negative, and so

$$\Delta H - T\Delta S < 0 \quad (2.2)$$

This implies that exothermic mixture ($\Delta H < 0$) will mix spontaneously, whereas for endothermic mixtures miscibility will only occur at high temperature. For two - component blends it is possible to construct a phase diagram, which may exhibit lower or upper critical solution temperature (LCST or UCST) (Figure 2.2). In

practice, LCST behavior is more commonly seen, phase separation occurring as temperature increases, because the intermolecular attractive forces responsible for the miscible behavior tend to disappear as the internal energy of the molecules becomes high enough to overcome them.

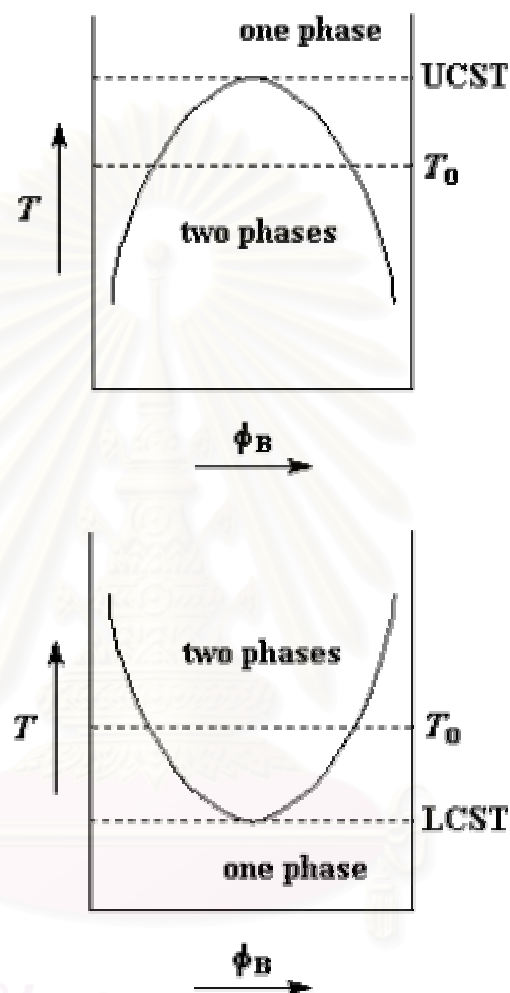


Figure 2.2 Schematic phase diagrams for binary blends showing LCST and UCST behavior[4].

b) Addition of Block and Graft Copolymers

The addition of block or graft copolymers represents the most extensively researched approach to compatibilization of blends. Block copolymers have been more frequently investigated than graft copolymer, and in particular block copolymer containing blocks chemically identical to the blend component polymers. It is perhaps not surprising that block and graft copolymers containing segments chemically

identical to blend components are obvious choices as compatibilizers, given that miscibility between the copolymer segments and the corresponding blend components assured, provided the copolymer meets certain structural and molecular weight requirements, and that the copolymer locates preferentially at the blend interfaces.

c) Addition of Functional/Reactive Polymers

Many workers have described the addition of functional polymers as compatibilizers. Usually a polymer chemically identical to one of the blend components is modified to contain functional (or reactive) units, which have some affinity for the second blend component; this affinity is usually the ability to chemically react with the second blend component, but other types of interaction are possible. The functional modification may be achieved in a reactor or via an extrusion-modification process[4]. Examples include the grafting of maleic anhydride or similar compounds to polyolefins, the resulting pendent carboxyl group having the ability to form a chemical linkage with polyamides via their terminal amino groups [6].

d) Reactive Blending

A comparatively new method of producing compatible thermoplastic blends is via reactive blending, which relies on the *in situ* formation of copolymers or interacting polymers. This differs from other compatibilization routes in that the blend component themselves are either chosen or modified so that reaction occurs during melt blending, with no need for addition of a separate compatibilizer. This route has found commercial application, for example in blends of polyamides with graft-functional polyolefin elastomers. Graft-functionalised elastomers, produced by melt modification, are commercially available for toughening nylons.

2.3 T_g- Composition Analysis of Miscible Polymer Blend

The T_g represents the temperature at which the polymeric chains have a combination of energy equal to the forces of attraction. Below this temperature, the polymer chains are locked into a random network with motion restricted to vibrational, rotational, and short order translational movement of small units of

polymer chain. Above this point, translational movement of the entire chain is possible, and diffusion therefore occurs as might be required. The observed T_g is affected by the frequency of the observed test method, pressure, crystallinity, and crosslinking.

Heterogeneous blends will exhibit distinctly different T_g 's as opposed to a homogenous blend with a single T_g . For miscible blends, T_g versus composition is not a universally similar relationship, but has many variations, similar to those observed with random copolymers. For polymer blends exhibiting miscibility over entire composition range, three generalized curves (in Figure 2.3) are possible: the linear relationship and the minimum and maximum deviation from linearity.

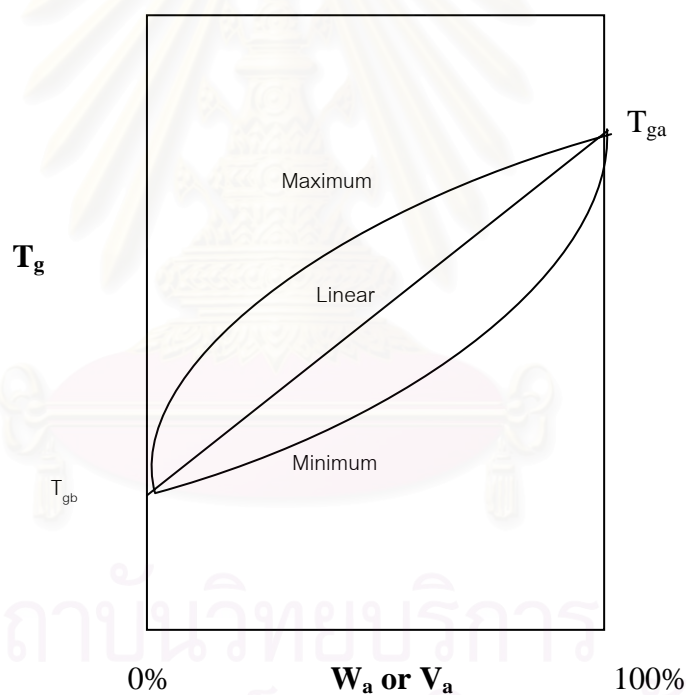


Figure 2.3 Generalized behavior of T_g relationships for miscible polymer blend [7].

In many cases, data fit relationships commonly used for random copolymers or for polymer-diluent blends, the Fox equation,

$$1/T_g = (W_a/T_{ga}) + (W_b/T_{gb}) \quad (2.3)$$

and the Gordon-Taylor equation,

$$T_g = [W_a T_{ga} + k(1-W_a)T_{gb}]/[W_a + k(1-W_a)] \quad (2.4)$$

where T_{ga} and T_{gb} represent the glass transition of the undiluted polymer components, W_a and W_b are the weight fractions of the blend, and k is the ratio of the thermal expansion coefficients between the rubber and glass states of the component polymer, $(\alpha_{1b} - \alpha_{gb})/(\alpha_{1a} - \alpha_{ga})$.

The Kelley-Bueche equation is similar to the Gordon-Taylor equation which expects that the volume fraction, ϕ_i , is used instead of the weight fraction:

$$T_g = [\phi_a T_{ga} + k(1-\phi_a)T_{gb}]/[\phi_a + k(1-\phi_a)] \quad (2.5)$$

As $\alpha_1 - \alpha_g$ has been proposed to be constant for all polymer, $k=1.0$ and the Gordon-Taylor and Kelley-Bueche equations reduce to the linear form:

$$T_g = W_a T_{ga} + W_b/T_{gb} \quad (2.6)$$

$$T_g = \phi_a T_{ga} + \phi_b/T_{gb} \quad (2.7)$$

The above equations, commonly used to express the T_g -composition relationships for copolymers and plasticizer-polymer compositions, are quite useful for miscible polymer blends [7].

2.4. Recycling Options for PET Bottles

Compared to glass, the advantages of PET bottles arise from their light weight and resistance to shatter, accompanied with further benefits in transportation and other related costs. These result, manufacturing practices for beverage bottles. On the other hand, PET degraded undergoes thermal and hygrothermal, which makes the problem when recycling via remelting and reduces the application potential of the recyclates. Its molecular weight is inevitably reduced during service and the major task to be solved with respect to recycling is increased, to restore the molecular weight. The goal behind this strategy should be the preferred operative route for any recycler.

2.4.1. Increase in Molecular Weight

Postcondensation processes usually enhance the molecular weight of PET. By operating PET in oxygen-free and moisture free environment at high temperatures, the

diffusion controlled process, this solid-state polycondensation. The chemical way is to use chain extending compounds that are able to react with functional group of PET in melt. As the reactivity of COOH and OH groups differ from one another even if the chain extender is able to react with both groups. The selection of chain extender should be based on the functional end groups attached tripolymeric chains in the melt, bifunctional molecules such as dianhydrides, diepoxides, and diisocyanates, turned out to be most suitable for the chain extension of PET [8-12].

Considering the type of functional groups in the chain extenders can easily strategy for reactive compatibilization of PET/polyolefin blends, the compatibilizer should have the same functional groups (anhydride, epoxy, and oxazoline) as chain extender.

2.4.2. Melt Blending

Major targets of PET blending were to improve the toughness and reduce the notch sensitivity of PET. The polymeric compatibilizers of polyester/polyolefin blends can be grouped into non-reactive and reactive polymers, depending on whether or not a chemical reaction between functional of the compatibilizer and polyester is excluded. For the compatibilization of immiscible blends, the traditional approach is to use preformed block or graft polymer, having affinity to both blend components. Their properties can not easily upon request and their use is further limited by the usually high price. The alternative is to generate such copolymers in situ by adding the functionalized polymer as the third component to the molten blend. The in situ produce compatibilizer is preferentially located at the interface and thus may efficiently lower the interfacial tension and promote the mechanical interlocking through entanglements between the originally immiscible phases. The most important interfacial chemical reactions for the praxis with polyesters are:

a) Hydroxyl (PET) + Anhydride or Acid (modifier). The reaction between the hydroxyl end group of PET and the anhydride or acid groups of functionalized polymer is still under dispute. This is due to the moderate reactivity of hydroxyl groups toward carboxylic groups and related anhydrides. Consequently, the improved compatibility between PET and acid or anhydride modified polymer is also argued to result from intermolecular dipole/dipole interactions and interchange

reactions between OH, COOH and ester groups [1]. The toughness improvement of PET, the favorite compatibilizers such as polymer or rubber grafted maleic anhydride, acrylic acid and succinic anhydride act as an emulsifier, reduce the interfacial tension and producing good adhesion between the blend components [14]. Acrylic acid grafted polypropylene (PP-g-AA) with and without additional transesterification catalysts was evaluated in a PET/PP blend obtaining 40 wt% PET [15], and maleic anhydride and acrylic acid in HDPE increase performance with PBT blend [16].

b) Carboxyl (PET) + Epoxy (modifier). The outstanding performance of PET/polyolefin blends compatibilized by epoxy-functionalized polymers can be attributed to the high reactivity of epoxy groups toward the carboxyl (and eventually hydroxyl) end-groups of PET. The grafting of GMA (glycidyl methacrylate) onto polymers in their melt. Bulk grafting of GMA may be accompanied by homopolymerization, resulting in poly(glycidyl methacrylate) (PGMA), an attempt was made to check whether PGMA is a suitable reactive modifier. PET blends with ethylene propylene rubber (EPR) and PGMA improve disperse size of polymer blends [17].

c) Carboxyl (PET) + Oxazoline (modifier). This reaction results in an esteramide linkage and thus can be used for the compatibilization of blends in which one of the polymers contains carboxyl groups, as it is the case with linear polyesters. Functional PP with ricinoloaxaline maleinate in the melt and used it for the compatibilization of PP/PBT blends [13] and oxazoline modifiers grafted on the phase structure of polyester/polyolefin blends are strong cohesion between the matrix and disperse phase [19].

d) Carboxyl/hydroxyl (PET) + Isocyanate (modifier). This reaction is evenly explored for the reactive modification of melt processable polyesters and their blends. The chemical reaction between the isocyanate group and the hydroxyl or carboxyl end groups of PET was traced by the increased compatibility. EPDM rubber grafted in solution with an isocyanate-contain monomer (reacting 2-hydroxyethyl methacrylate with isophorone diisocyanate) can well be dispersed in PET [20].

e) Carboxyl (PET) + Nitrile (modifier). This reaction results in imide moieties and was explored for the toughness improvement of PBT by adding NBR

[21], this reaction may also occur in linear polyesters blended with acrylonitrile-butadiene-styrene (ABS) [22].

f) Transesterification Reaction (between PET and modifier). Though this strategy is extensively followed in polyester blends with other polycondensates, in this case of PET/polyolefin blend, it is used to a lesser extent. Diethylmaleate grafted PEs (PE-g-DEM) proved to be suitable modifiers for PET/LLDPE blends.

2.4.3 Thermoplastic Elastomers

The reactive modification of linear polyester with various elastomers did not always follow the alternative routes listed above. The epoxied EPDM was used to modified polyester compound. Copolyesters containing diethyl maleate units were thought to react EPDM via a reaction resulting in a finer dispersion of EPDM [23]. Reactions between macromolecules, in the presence of bismaleimide were also claimed to improve to compatibility between PBT and EPDM [24]. Thermoplastic elastomers containing PBT and EPDM-g-GMA in 50/50-wt%, the dispersed rubber component was crosslinked via peroxide or with diamine compounds that reacted with the residual epoxy groups of the EPDM-g-GMA. The structure of dynamic vulcanizates is a continuous thermoplastic matrix where the rubber is dispersed in small-vulcanized particles. The blends with a stable co-continuous phase morphology often referred to as interpenetrating network (IPN) structure. The advantage of this IPN structure is that undesirable effects due to the varying quality of the PET scar or physical aging are much better tolerated by this morphology than by a dispersed one.

The compatibility of PET blends with GMA-grafted rubbers was improved with the epoxy-function.

Functionalized rubber is partly replaced by acrylonitrile butadiene rubber (NBR) of high acrylonitrile content. The dynamic vulcanization of the customer phase by peroxide had only a marginal effect on the mechanic and thermomechanical performance of the blend [25].

2.5. Ethylene-Propylene Rubber Modification.

In recent years, an increasingly important application of EPDM has been in blending with plastics to obtain enhanced material properties. Because of their compatibility, EPDMs and EPDMs are particularly suited for blending with polypropylene and polyethylene. There are two classes of plastic/rubber blends. TPOs (thermoplastic olefins) and TPEs (thermoplastic elastomers) are essentially rigid at the service temperature but the brittleness has been reduced by the addition of EPDM to produce a soft plastic, thus increasing their utility. TPEs exhibit rubbery properties at the service temperature but will process as thermoplastics. Both classes of polymer have found their major application to date in automotive applications such as bumper covers, decorative facial, and air ducting; however, wire and cable and hose applications are showing substantial rates of growth.

Introducing succinic groups (SAH) in ethylene-propylene (EPM) rubbers is a useful method for improving their adhesion characteristic and for obtains new materials designed for applications, depending on the elastomer's end use. Two main synthetic routes have been used to produce EPDM-g-SAH copolymers: solution and bulk techniques.

2.5.1 Chemical Modification of Ethylene-Propylene Rubber

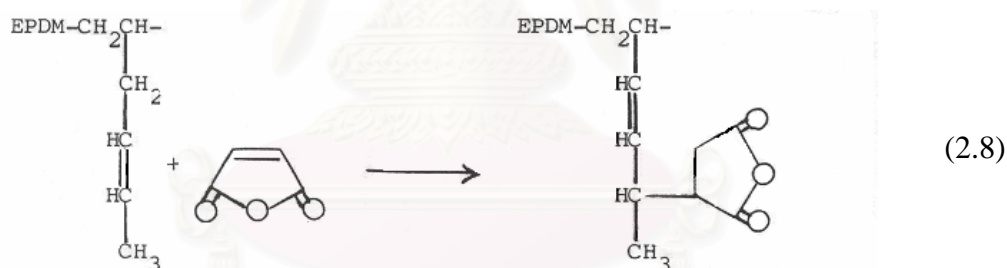
Chemical modification technique of ethylene propylene rubber, solution process was studied early, probably because elastomer solution produced by Ziegler-Natta catalyzed synthesis could be applied directly[25]. The reactants' and products' solubility in aromatic hydrocarbons made them a natural choice of solvents for grafting EPM with maleic anhydride (MAH) and derivatives [26]. Peroxide initiators are used the most for grafting MAH and its derivatives onto the elastomer backbone. Monomer reactivity played a decisive role when solvent influence was evaluated. Solvents with higher chain transfer constants reduced the yield of elastomer chemical modification. Using the solvent with higher chain transfer constant, as well as a decreasing peroxide content in the reaction medium resulted in reduced EPM degradation.

The preferred way of introducing MAH and its derivatives in DPDM elastomers is by bulk technique, continuous process for modifying EPDM with MAH

with extruder as polymerization reactors, to obtain homogenous reaction conditions with relatively low barrel temperatures. The grafting efficiency was mainly determined by the initiator: MAH ratio, and an expected decrease in the amount of grafted MAH with the increase of MAH content in the charge was verified. Reactions can be carried out with or without peroxide initiators. When this initiator are used, they do not only degrade the base rubber, but also cause elastomer crosslinking [26].

2.5.2 Mechanism Features

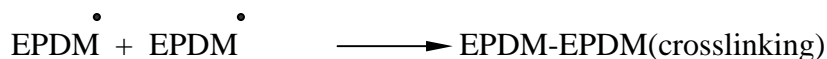
The mechanism of the grafting reaction in the presence of peroxides is still not clear. Two different mechanisms have been proposed for grafting MAH onto polyolefinic substrates. The conventional mechanism involves only free radical reactions and cannot explain satisfactorily the variation of reaction behavior depending on MAH content in the reaction medium. The mechanisms explain the effect of charged MAH on the side reaction. Unusually low grafting efficiencies and high oxidation indexes were obtained for formulations with a high MAH content. According to the mechanism, the following reactions may take place [26].



And side reaction is as follows:



or



2.6 Literature Reviews

Papke, et al. [25] studied thermoplastic elastomers based on compatibilized poly(ethylene terephthalate) blends: effect of rubber type and dynamic curing. Thermoplastic elastomers containing poly(ethylene terephthalate) (PET) in 50wt%, compatibilizer (glycidyl methacrylate grafted rubber or glycidyl methacrylate containing copolymer) in 30wt% and various rubbers (20wt%) were produced by melt blending with and without dynamic curing (dicumyl peroxide initiated). The static tensile (stress-strain behavior) and dynamic mechanical thermal properties (DMTA) of the systems along with their phase morphology (scanning electron microscopy of cryofractured and etched surfaces) were determined. It was found that the blend compatibility with PET is strongly improved when a high acrylonitrile- containing nitrile rubber (NBR) and an ethylene-glycidyl methacrylate copolymer (EGMA) or a GMA grafted ethylene/propylene rubber (EPR-g-GMA) are used as rubber and/or compatibilizer in the blend recipes. The effect of dynamic curing on the tensile and DMTA properties of the blends was negligible. Fractographic inspection of the fractured surface showed the development of a co-continuous phase structure which was in accordance with learnings from the DMTA spectra.

Sanchez-Solis, et al. [27] studied influence of maleic anhydride grafting on the rheological properties of poly(ethylene terephthalate)-styrene butadiene blends. The rheological properties of poly(ethylene terephthalate) (PET)-styrene butadiene rubber (SBR) blends are studied in this work. The shear viscosity was measured in the special capillary device mounted on an injection-molding machine, which provides data on pressure and flow rate. Comparison of viscosity of PET-SBR physical blend with that of the blend of PET with maleic anhydride-functionalized SBR (SBRg) provides information on the effect of grafted maleic anhydride chains at the rubber-thermoplastic interface on the rheological properties of the blend. Shear viscosity is a function of the amount of compatibilizer and applied shear stress present in the preparation of the blends. A reduction in the particle size due to increasing screw speed was correlated with a reduction in the shear viscosity and an increase in the impact properties of the blend.

Jin-Gyu, et al. [28] studied blends of poly(ethylene terephthalate) with EPDM through reactive mixing. Ethylene-propylene diene elastomer (EPDM), which is grafted with an isocyanate-containing monomer (HI), was blended with poly(ethylene terephthalate) (PET) and its morphological, thermal, rheological, and mechanical properties were studied. HI was incorporated onto EPDM backbone through a solution graft reaction. When the PET was blended with HI-grafted EPDM (EPDM-g-HI), the morphologies of dispersed phases showed considerable differences in the aspects of particle size and interfacial adhesion compared with those of PET/EPDM blend. DSC analysis showed that, when blends are cooled slowly, the PET phase in PET/EPDM-g-HI is somewhat amorphous compared with that in the PET/EPDM blend. The increase in complex viscosity, storage modulus and impact strength of PET/EPDM-g-HI blends enabled us to ensure that the compatibility between PET and EPDM improved through functionalization of EPDM with the isocyanate moiety. These results are mainly due to graft PET-EPDM copolymer in situ formed through the chemical reaction between the isocyanate group of EPDM-g-HI and hydroxyl or carboxyl end groups of PET.

Sanchez-Solis, et al. [29] studied on the properties and processing of poly(ethylene terephthalate)/styrene-butadiene rubber blend. The mixing of incompatible polymers such as poly(ethylene terephthalate) (PET) and styrene-butadiene rubber (SBR) produces a blend with poor mechanical and impact properties, because polymeric phase interact weakly with each other and segregate. The use of SBR grafted with maleic anhydride (MAH) increases the compatibility of SBR-PET system by generating higher interactions and chemical links between the ingredients of the blend. The induced compatibility is reflected in the 2.5-fold increase in the impact resistance of the blend as compared to that of pure PET. The grafting reaction to produce SBR-g-MAH is carried out by reactive extrusion using a reaction initiator, benzoyl peroxide (BPO), and the extent of the reaction depends on the content of MAH and BPO. Results indicate the close relationship between processing conditions and microstructural parameters, such as particle diameter and interparticle distances of the dispersed rubber phase, necessary to achieve the optimum impact resistance.

Mouzakis, et al. [17] studied fracture toughness assessment of poly(ethylene terephthalate) blends with glycidyl methacrylate modified polyolefin elastomer using essential work of fracture method. The static fracture toughness of poly(ethylene terephthalate) (PET) melt blended with a modifier containing glycidyl methacrylate (GMA)-grafted ethylene propylene rubber and homopolymerized GMA were studied on injection molded specimens by adopting the essential work of fracture (EWF) method. It was found that the essential and nonessential or plastic work both decrease with an increasing amount of modifier (up to 20 wt%) if the PET matrix is amorphous and nonaged. The scatter in the EWF data for the blend with 10 wt% modifier was found by presuming concurrent mechanism between microcrystallization and morphology-dependent cavitation and fibrillation processes.

Blow, et al. [30] studied the modification of isotactic PP/HDPE blend with EPDM and EVA. An addition of high-density polyethylene (HDPE) to PP resulted in a significant reduction in the elongation at break and in the gate region impact strength of injection molded articles. The compatibilizers employed in their study were ethylene/propylene/diene monomer (EPDM) or EVA copolymers. The former improved effectively the impact strength and gate puncture resistance of the PP/HDPE blend weight ratio of 90/10. EVA, on the other hand, improved effectively the impact strength and the elongation at break of the polyblend. It was found that EVA was better impact modifier for PP than EPDM. DSC analysis indicated that EPDM blended with PP caused a significant reduction in the degree of crystallinity of the PP. This was not observed in the case of EVA/PP blends.

Sanchai, et al. [31] studied ethylene-vinyl acetate copolymer impact modifier for polypropylene. The study of utilization of ethylene-vinyl acetate copolymer (EVA) having 28% by weight of vinylacetate (VA) content as an impact modifier for isotactic polypropylene (PP) was carried out as well as the study of a possibility to use polypropylene grafted maleic anhydride (PP-g-MA) as a compatibilizer. Effect of screw speed of a twin screw extruder, amount of dicumyl peroxide and maleic anhydride (MA) on the percentage of MA grafting onto PP and melt flow index of resulting PP-g-MA were studied. The optimum condition for the preparation of PP-g-MA was found, the screw speed should be of 90 rpm, dicumyl peroxide of 4 phr, and MA of 6.3 phr. The properties of resulting PP-g-MA were 0.7 %grafting, and melt

flow index of 25 g/10min. This PP-g-MA was employed as the compatibilizer for PP/EVA blend which EVA in the polyblend were in the range of 0-50% and PP-g-MA of 0-10 phr. EVA having 28% VA content was better impact modifier than that having lower VA content. Usage of 2 phr PP-g-MA slightly improved the impact strength property. Morphology of the polyblend composed of EVA droplet dispersed in the matrix of PP. When compared with other elastomer, EVA was better modifier than the others, except styrene-ethylene-butadiene-styrene (SEBS) at 20%. Tensile strength at yield and hardness of polyblend decreased with the increase in EVA amount and the usage of PP-g-MA did not affect on that properties. Elongation at break of polyblend increased with the increase in EVA content in polyblend from 5 to 40%. If the amount was over than 40%, this property was poorer. The usage PP-g-MA slightly decreased the elongation at break when the EVA content in polyblend was 5-30%. The large decrease of this property was when the EVA content in the polyblend was upto 40-50%, especially, when the amount of PP-g-MA used was 6 and 10 phr. For the miscibility of polyblend, was found that EVA could be only miscible with the amorphous region of PP.

CHAPTER III

EXPERIMENTAL

3.1 Materials

PET pellet bottle grade supplied by Bangkok Polyester Co., Ltd. (I.V. 0.80 dl/g). EPDM grade Buna3950 (Ethylene content = 56 ± 4 , EBN content = 11.4 ± 1.4 and Mooney viscosity (1+8) $100^\circ\text{C} = 54 \pm 6$) and Buna6740 (Ethylene content = 68 ± 4 , EBN content = 4.5 ± 0.6 and Mooney viscosity (1+8) $125^\circ\text{C} = 55 \pm 5$) were supplied by Bayer Co., Ltd.

Benzoyl peroxide (BPO) and maleic anhydride (MAH) supplied by Merck Co., Ltd. were used as received.

3.2 Instruments

The major instruments used in this study are listed as follows:

1. Single screw extruder (Model BX 18, Axon ab)
2. Pendulum impact testing machine (Model 6546, Ceast)
3. Universal testing machine (Model H 50 KS, Hunsfield)
4. Compression (Model LP 50, Labtech Engineering)
5. Differential scanning calorimeter (Model DSC7, Perkin-Elmer)
6. Melt Flow Index (Model 6542, Ceast)
7. Scanning Electron Microscope (Model JSM-5800LV, JEOL)

3.3 Experimental

3.3.1 Grafting Maleic Anhydride onto EPDM

500 g EPDM pellet (Buna6740 and Buna3950), fixed amount of BPO at 0.15 phr and varied amount of MAH 0, 1, 3 and 5 phr were mixed in a plastic bag for 10 minutes. At mixed materials were subsequently fed into a hopper of the single screw extruder which the temperature of four heating zones were controlled at 150°C, 180°C, 180°C, 190°C and die zone was at 180°C. Screw speed of single screw extruder employed in the experiments was 30 rpm. The long strand extrudates were chopped into granules using the palletizer.

Table 3.1 Quantities of BPO and MAH for the preparation of maleic anhydride grafted EPDM (EPDM-g-MAH)

	Buna6740 (g)	Buna3950 (g)	MAH (phr)	BPO (phr)
B3950	1000	-	0	0.15
B3950-g-1MAH	1000	-	1	0.15
B3950-g-3MAH	1000	-	3	0.15
B3950-g-5MAH	1000	-	5	0.15
B6470	-	1000	0	0.15
B6470-g-1MAH	-	1000	1	0.15
B6470-g-3MAH	-	1000	3	0.15
B6470-g-5MAH	-	1000	5	0.15

3.3.2 Preparation of PET and EPDM (PET/EPDM-g-MAH) Blends

The total amount of 2000 g of PET and EPDM were used for the preparation of PET/EPDM blend. The amount of EPDM in polymer blend varied from 20% and 30% by weight. Each of the polymer blend composition, the MAH grafted onto EPDM was varied in the range of 1 to 5 phr.

The effects of various MAH contents grafted onto EPDM, EPDM composition and method of blending for PET/Buna3950, PET/Buna6740 and recycled PET/

Buna3950 blends were studied. The blending ratios of PET/EPDM blends are presented in Table 3.2.

Table 3.2 Blending ratio of PET/Buna3950, PET/Buna6740 and recycled PET/Buna3950.

	Ratio	MAH grafted onto EPDM *(phr)	MAH grafted onto EPDM **(phr)
PET/Buna3950	70/30	0	-
	70/30	3	-
	70/30	-	3
	80/20	0	-
	80/20	1	-
	80/20	3	-
	80/20	5	-
	80/20	-	3
PET/Buna6740	80/20	0	-
	80/20	1	-
	80/20	3	-
	80/20	5	-
	80/20	-	3
Recycled PET/Buna3950	70/30	3	-
	80/20	3	-

*Two-step blending = Grafting of MAH onto EPDM before blending with PET.

**One-step blending = Grafting of MAH onto EPDM during blending with PET.

Typically, all of materials mentioned above were hand-mixed in a plastic bag by tumbling up and down for 30 minutes. The mixed material was then introduced into a hopper of the single screw extruder, with the controlled temperature of three heating zones at 220°C, 240°C, 240°C, 250°C and die zone was at 240°C. The screw speed was fixed at 50 rpm. The long strand extrudates were chopped into granules using the pelletizer .

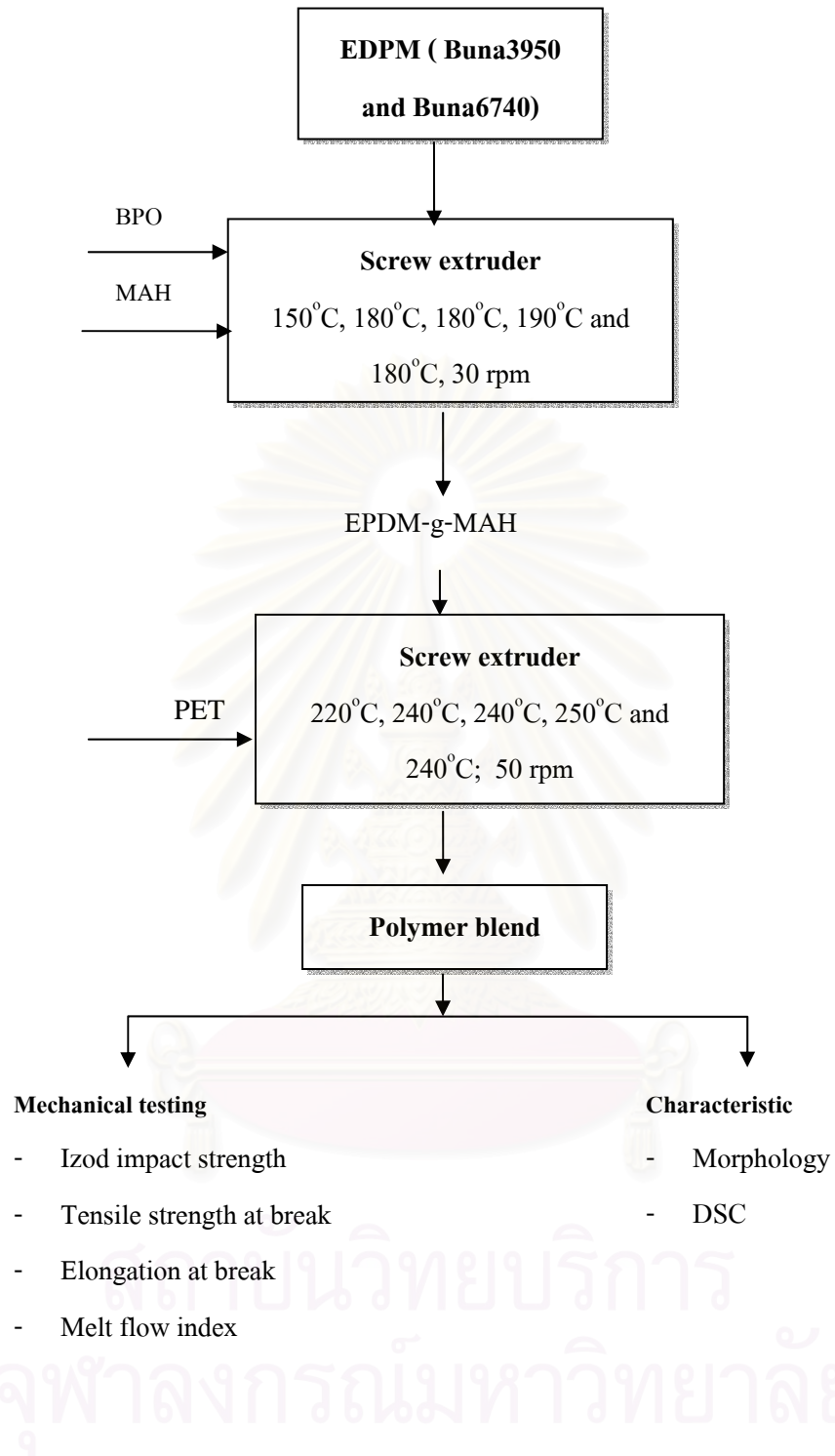


Figure 3.1 Diagram of experimental procedure to prepare EPDM-g-MAH and PET/EPDM blends.

3.4 Characterization of EPDM-g-MAH

The reacted MAH presented in EPDM-g-MAH granulates was determined by measuring the acid number by following procedure: One gram of grafted rubber was dissolved into 100 ml toluene at 65°C for 10 hours. 50 ml of water were added into the solution and then the solution was separated into three phases: organic phase, gel and aqueous phases. A solution of organic phase was taken and titrated with 0.005 N ethanolic KOH using phenolphthalein as an indicator. The end point could be observed by colour change from colourless to pink solution [35]. The experiments were repeated at least two times. The weight percentage of MAH grafting on EPDM was calculated by equation (3.1). EDPM that used as the starting materials for preparation of EDPM-g-MAH was used as a blank following the method described above. Therefore, the average value of MAH content in the grafted polymer repeated subsequently were those already subtracted by the blank values.

$$\% \text{ MAH grafting onto EPDM} = \frac{(\text{mole of KOH}) \times 98 \text{ g-mol}^{-1} \times 100}{2 \times \text{weight of sample (g)}} \quad (3.1)$$

3.5 Melt Flow Index (MFI)

MFI measurement was carried out on Melt Flow Index (Model 6542, Ceast) at 285°C with the hammer load of 2.16 kg in accordance with ASTM 1238. Seven samples of each blend were measured. The average values of five samples between the maximum and minimum values were taken.

3.6 Mechanical Properties

3.6.1 Izod Impact Strength Testing

Test specimens for the measurement of Izod impact strength were prepared following ASTM D4101. They were tested according to the standard method of ASTM D256. Seven samples of each blend were measured. The average values of five samples between the maximum and minimum values were taken.

3.6.2 Tensile Property Measurement

The dumbbell specimens for the tensile property measurement were prepared according to ASTM D4101. They were tested in accordance with ASTM D638. Seven samples of each blend were measured. The average values of five samples between the maximum and minimum values were taken.

3.7 Differential Scanning Calorimetric Technique

25 mg sample was heated from 30 to 270°C under nitrogen atmosphere at a heating rate of 200°C/min, and held for 10 min. The sample was then cooled from 200 to 30°C at 10°C/min and hold for 5 min. After that, the sample was heated from 30 to 270°C at 10°C/min. Data were collected during the second heating cycle.

3.8 Morphology of Polymer Blends

Scanning electron microscopy (SEM) was used to observe the morphology of the polymer blends. The specimens were immersed in liquid nitrogen for 30 min, then were broken rapidly. The broken surface was coated with platinum-palladium using T102 Hitachi coating unit.

CHAPTER IV

RESULTS AND DISCUSSION

4.1 Grafting of Maleic Anhydride onto Ethylene Propylene Diene Elastomer.

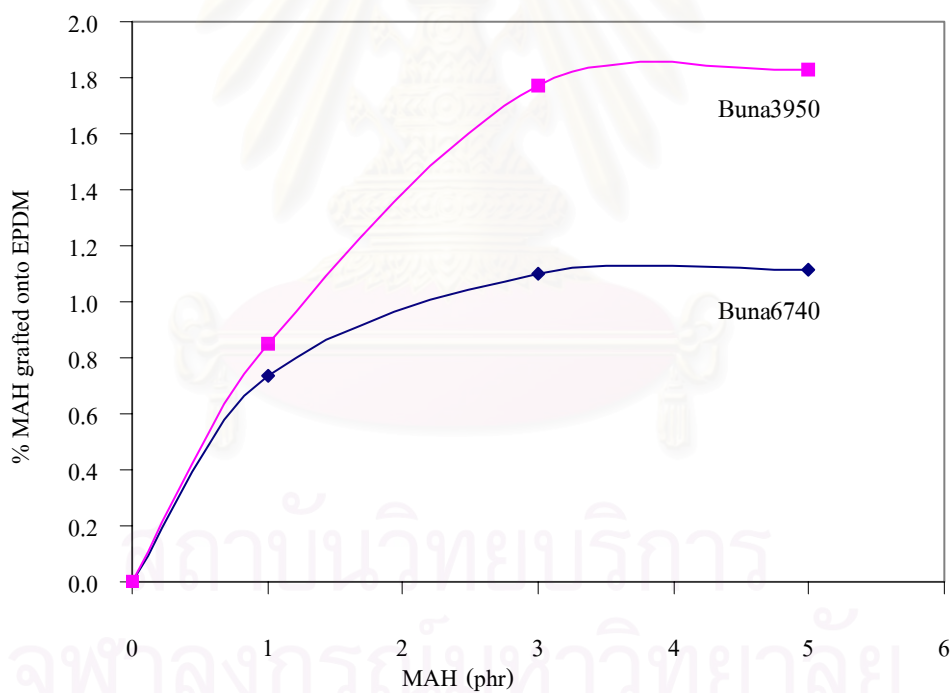
In this work, the effect of maleic anhydride content grafted onto two types of EPDM at various contents was studied. After MAH being grafted onto EPDM, MAH was separated from EPDM-g-MAH by extraction. The amount of MAH grafted onto EPDM was determined by titration and calculation (see the results in Appendix A). The results of variation of MAH content at constant initiator (BPO) content of 0.15 phr and screw extruder speed of 30 rpm are shown in Table 4.1.

Figure 4.1 shows the grafting degree at a BPO content of 0.15 phr for initiated grafting reaction MAH onto EPDM via a single screw extruder at 180°C. The result is obvious that % MAH grafted onto EPDM increases with increasing MAH content in the range of 0 to 3 phr for both EPDM types. Further increase of MAH content upto 5 phr, there is no significant effect of % MAH grafted onto EPDM. Since the melt of MAH and EPDM was immiscible, the grafting reaction occurred within limited interfacial region. So the efficiency of grafting depends on the rate of MAH diffusion into EPDM phase. And rate of MAH diffusion into EPDM is likely increased with an increasing MAH content. An increasing MAH content leads to a steep increasing grafting degree. Until the one content reached, the grafting degree does not increase but there is increased in crosslinking [32].

Comparison between Buna3950 and Buna6740, % MAH grafted onto Buna3950 were higher than that onto Buna6740 for every MAH content used. This could be explained by the structure of Buna3950 (EBN content 11.4%) contain diene amounts more than that of Buna 6470(EBN content 4.5%), therefore, Buna3950 could be reacted with MAH more than Buna 6470 [6].

Table 4.1 Grafting degree of EPDM-g-MAH at various MAH contents.

Elastomer type	MAH (phr)	BPO (phr)	% MAH grafted on EPDM
Buna3950	1	0.15	0.86
	3	0.15	1.79
	5	0.15	1.83
Buna6740	1	0.15	0.74
	3	0.15	1.10
	5	0.15	1.11

**Figure 4.1** Effect of MAH content on grafting degree of EPDM-g-MAH.

4.2 Melt Flow Index of PET/EPDM-g-MAH Blends

The effect of MAH content and diene content of EPDM on melt flow index was studied. The melt flow index of PET/EPDM-g-MAH at 80/20 ratio with variation of MAH content are presented in Table 4.2 and Figure 4.2

Upon maintaining the ratio of the blends at 80/20 and variation of the MAH content, it was found that the % MAH grafted onto EPDM increased with increasing MAH content, obviously resulted in the decreasing melt flow index. The EPDM grafted with MAH dispersed and acted as compatibilizer in the blends. The MAH reacted to hydroxyl (OH), carboxyl (COOH) and ester functional group in PET chain and EPDM segment in EPDM-g-MAH entangled in EPDM phase that lead to cohesion between PET and EPDM phase. This also caused an increase in viscosity of the blends.

Table 4.2 also presents the comparison of melt flow index between PET/Buna 3960-g-MAH and PET/Buna6740-g-MAH blends. Melt flow index of PET/Buna 3960-g-MAH blends was lower than that of PET/Buna6740-g-MAH, because the mooney viscosity of Buna3950 was lower than that of Buna6740. Therefore, this is the direct effect on melt flow index of the blend. Another reason is the difference in diene content of Buna3950 and Buna6740. The Buna3950 contains more diene content than Buna6740, therefore %MAH grafted onto Buna3950 was higher than that onto Buna6740(see Table 4.1). This affects the efficiency of cohesive interaction between the MAH grafted on EPDM and the functional group in PET chain.

Table 4.3 shows the melt flow index of PET/Buna3950-g-MAH blend at 70/30 and 80/20 ratio for various blending methods. An increase in EPDM content of blend decreased the melt flow index because the viscosity of EPDM is higher than that of PET. This can be explained that EPDM dispersed and entangled in PET phase that lead to the decrease in melt flow index.

The grafting of MAH onto EPDM before blending with PET (two-step blending) and the MAH grafting during blending of PET/EPDM (one-step blending) were studied at 3 phr of MAH content. The results show that melt flow index of two-step blending was lower than that of one-step blending because for the one-step blending, MAH could reacted with both EPDM and PET. It can be assumed that some

amount of MAH diffused to react with functional groups of PET in the short time. This leads to the loss of MAH content to graft onto EPDM, so the amount of grafted EPDM in the two-step blending was higher than that in the one-step blending. This is the consequence of the interaction between distributed EPDM phase containing grafted EPDM and PET phase.

Table 4.2 Melt flow index of PET/EPDM-g-MAH blends with various MAH contents for PET/EPDM blends at 80/20 ratio.

Ratio	Melt Flow Index (g/10 min)	
	PET/Buna6740	PET/Buna3950
100/0	63.1	63.1
80/20	60.7	53.2
80/20-g-1*	60.2	48.6
80/20-g-3*	59.1	45.6
80/20-g-5*	58.9	44.2
80/20/3**	60.5	53.1

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

Table 4.3 Melt flow index of PET/Buna3950-g-MAH blends at 3 phr MAH and various PET/EPDM blend ratios.

Compatibilizer	Melt Flow Index (g/10 min)	
	PET/Buna3950-g-MAH at 80/20	PET/Buna3950-g MAH at 70/30
	0	60.7
Buna-g-MAH 3 phr (Two-step blending)	59.1	40.3
MAH 3 phr (One-step blending)	60.6	45.2

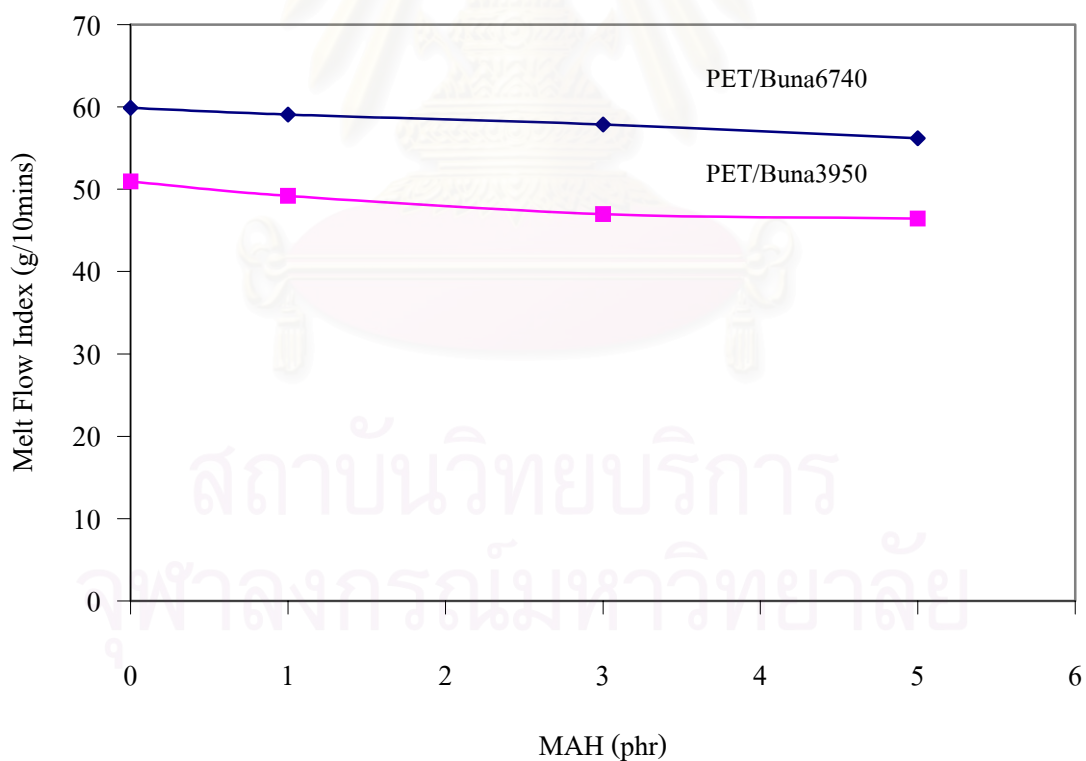


Figure 4.2 Effect of MAH content on melt flow index of PET/EPDM-g-MAH blends.

4.3 Mechanical Properties of PET/EPDM-g-MAH Blends

4.3.1 Impact Strength of PET/EPDM-g-MAH Blends

Table 4.4 shows the impact strength of PET/EPDM blends with and without MAH grafted onto EPDM at 80/20 blend ratio. Figure 4.2 compares the impact strength of both PET/EPDM (Buna3950 and Buna6740) blends. MAH in the range of 0 to 5 phr was employed as the compatibilizer via grafting onto EPDM in preparation of PET/EPDM-g-MAH blends. The results show that when the MAH content in EPDM increased, their impact strength was not much improved. Significant improvement was found at 3 phr MAH grafted onto EPDM.

The impact strength of the polymer blends depends on dispersed particle size and interfacial adhesion. As can be seen in Figures 4.10-4.21 (SEM micrographs), it is clear that impact strength of the blends is dependent strongly upon the blend morphology. When EPDM was blended in PET, the dispersed EPDM could not act as an effective stress dispersant due to large dispersed phase size. However, when EPDM-g-MAH was blended with PET, the dispersed EPDM phase became small enough to act as stress dispersant. Moreover, the partial adhesion between the phase would transfer the stress field from the PET matrix to dispersed elastomer phase effectively.

The impact strength for 80/20 and 70/30 blend ratios of PET/Buna3950-g-MAH blends is presented in Table 4.3. It is apparent that the higher EPDM content (70/30 ratio) showed the better impact strength than the blends that have lower EPDM content (80/20 ratio). There was dependence of impact strength on elastomer content. Thus, increasing EPDM content in blends causes the increase in transferring stress under impact force.

For the two blending methods, the blending of EPDM-g-MAH and PET (two-step blending) showed the higher impact strength than the blending of MAH, EPDM and PET (one-step blending). This can be explained that the diffusion of MAH grafted onto EPDM in two-step blending is higher than that of one-step blending.

Tables 4.6 and 4.7 also show the correlation of relative impact strength of PET/EPDM-g-MAH blends to the MAH content in the range of 0 to 5 phr. The relative impact strength is defined as follows:

$$\text{Relative impact strength} = \frac{\text{Impact strength of PET/EPDM blend}}{\text{Impact strength of PET}} \quad (3.1)$$

The impact strength of PET was 11.97 J/m.

From Table 4.6, at 80/20 blend ratio, EPDM with 3 phr MAH seemed to be good candidate for impact strength improvement of PET, relative impact strength for PET/Buna6740 and PET/Buna3950 are 1.21 and 1.41, respectively. Moreover, at the 70/30 blend ratio of PET/EPDM blend with grafted MAH of 3 phr, impact strength was improved approximately 2.8 times pure PET.

Table 4.4 Impact strength of PET/EPDM-g-MAH blends at various MAH contents for PET/EPDM blend ratio of 80/20.

Ratio	Impact strength (J/m)	
	PET/Buna6740	PET/Buna3950
100/0	11.9	11.9
80/20	12.3	11.9
80/20-g-1*	13.5	12.0
80/20-g-3*	14.5	16.9
80/20-g-5*	13.5	11.9
80/20/3**	12.5	10.3

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

Table 4.5 Impact strength of PET/Buna3950-g-MAH blends at 3 phr MAH and various PET/EPDM blend ratios.

Compatibilizer	Impact strength (J/m)	
	PET/Buna3950-g-MAH at 80/20	PET/Buna3950-g MAH at 70/30
	0	12.3
Buna-g-MAH 3 phr (Two-step blending)	14.5	33.6
MAH 3 phr (One-step blending)	12.5	31.1

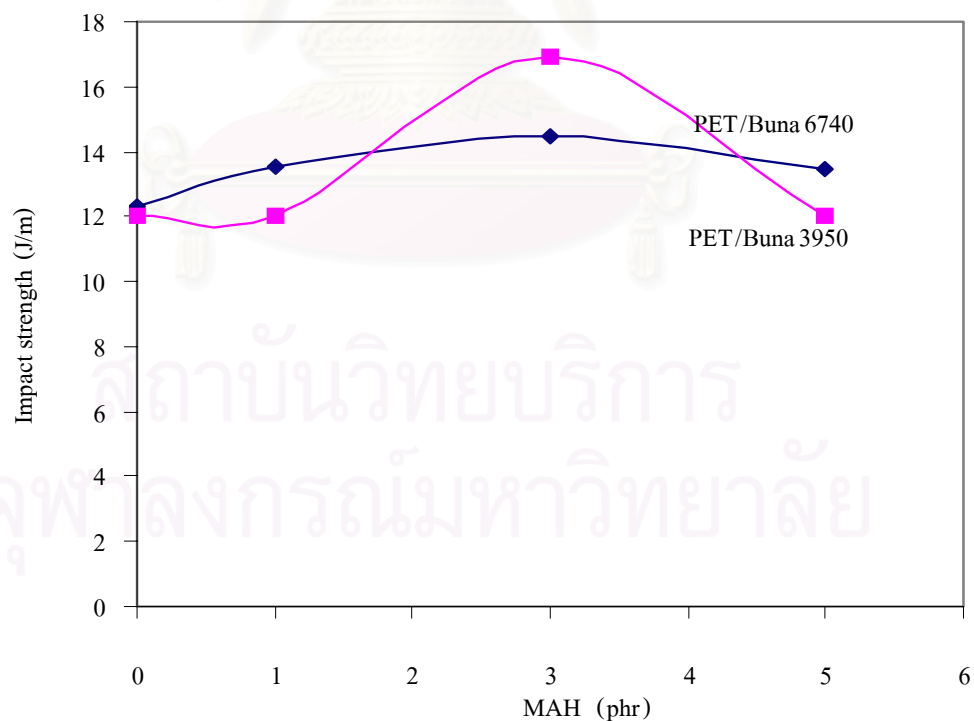


Figure 4.3 Effect of MAH content on impact strength of PET/EDPM-g-MAH blends.

Table 4.6 Relative impact strength of PET/EPDM-g-MAH blends at various MAH contents for PET/EPDM ratio of 80/20.

Ratio	Relative impact strength	
	PET/Buna6740	PET/Buna3950
80/20	1.03	1.00
80/20-g-1*	1.13	1.00
80/20-g-3*	1.21	1.41
80/20-g-5*	1.13	1.00
80/20/3**	1.04	0.86

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

Table 4.7 Relative impact strength of PET/Buna3950-g-MAH blends at 3 phr MAH and various PET/EPDM blend ratios.

Compatibilizer	Relative impact strength	
	PET/Buna3950-g-MAH at 80/20	PET/Buna3950-g MAH at 70/30
0	1.03	1.14
Buna-g-MAH 3 phr (Two-step blending)	1.21	2.81
MAH 3 phr (One-step blending)	1.04	2.59

4.3.2 Tensile Strength of PET/EPDM-g-MAH Blends

Table 4.8 and Figure 4.4 show the tensile strength of PET/EPDM blends at blend ratio of 80/20 with and without MAH grafted onto EPDM as compatibilizer. Effect of MAH content in PET/EPDM-g-MAH blends was studied. Tensile strength of the blend without grafted MAH was lower than that of the original PET. After addition of MAH graft onto EPDM, tensile strength of PET/EPDM showed increasing trend according to increasing amount of MAH in PET/EPDM-g-MAH blends. The improvement in tensile strength is similar trend of the impact strength that can be attributed to change of their morphologies and improve interfacial adhesion. Tensile properties of immiscible and partially miscible blends depend on the adhesion between two phases and domain size of dispersed component, both are controlled mainly by the interfacial tension. The results of scanning electron microscope also supported the tensile strength results (see Figures 4.10-4.21). The blend with MAH content had finer phase domain size than the blend without MAH grafting on EPDM. An effective compatibilizer preferably resided along the interface.

For the comparison of PET/Buna3950-g-MAH and PET/Buna6740-g-MAH, tensile strength of the blend increased with increasing MAH content and both EPDM types exhibited similar behavior. Table 4.9 shows the change of tensile strength with EPDM content in the blends. Tensile strength of PET/EPDM blends decreases with increasing EPDM content, because EPDM has lower tensile strength than PET. It can be concluded that the tensile strength of the blend depends on the morphology of matrix phase (PET) and average size of dispersed phase.

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Table 4.8 Tensile strength of PET/EPDM-g-MAH blends at various MAH contents for PET/EPDM ratio of 80/20 .

Ratio	Tensile strength (MPa)	
	PET/Buna6740	PET/Buna3950
100/0	24.45	24.45
80/20	9.207	9.16
80/20-g-1*	11.55	11.45
80/20-g-3*	14.06	12.86
80/20-g-5*	15.15	15.22
80/20/3**	7.45	8.24

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

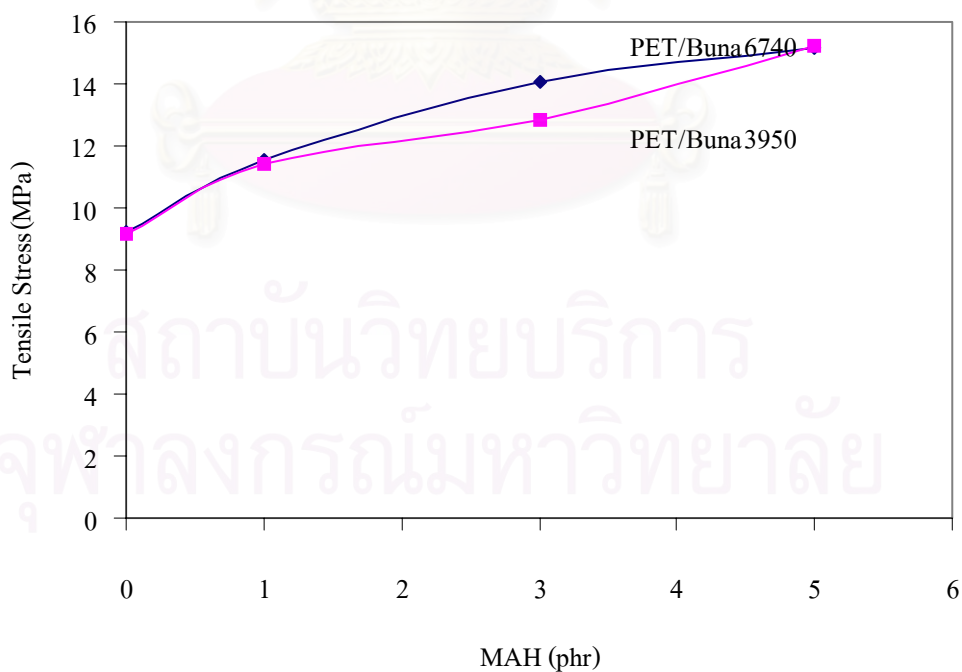


Figure 4.4 Effect of MAH content on tensile strength of PET/EDPM-g-MAH blends.

Table 4.9 Tensile strength of PET/Buna3950-g-MAH blends at 3 phr MAH and various PET/EPDM blend ratios.

Compatibilizer	Tensile strength (MPa)	
	PET/Buna3950-g-MAH	PET/Buna3950-g MAH
	at 80/20	at 70/30
0	9.21	6.28
Buna-g-MAH 3 phr (Two-step blending)	14.06	6.06
MAH 3 phr (One-step blending)	7.45	5.58

The effect of melt grafting on PET/EPDM-g-MAH blends was studied by comparison of two blending methods and two PET/EPDM ratios (see Table 4.9). The tensile strength of blends by grafting of MAH on EPDM before blending with PET (two-step blending) is higher than that of blending by one-step blending. Using one-step blending method, some of MAH may react with PET in chain scission reaction and this may possibly cause the lower tensile strength. From Table 4.9, the tensile strength of PET/EPDM at ratio of 70/30 is lower than that of blend at ratio of 80/20. This is caused by the low tensile strength of EPDM.

4.3.2 Elongation at Break of PET/EPDM-g-MAH Blends

Effect of MAH content grafted of PET/EPDM blends on the elongation at break is shown in Table 4.10 and Figure 4.5. From these results, the blends without MAH graft onto EPDM showed the lower elongation at break than pure PET. After addition of the various MAH contents onto EPDM, the elongation at break was improved from the blend without compatibilizer and also higher than that of pure PET. The compatibilizer improved the adhesion between two phases and stabilized elastomer phase in polymer blend. The effect on polymer blend is achieving better elongation than without MAH.

From Tables 4.10 and 4.11, elongation at break of the blends does not change when EPDM content increased. In general, PET property is brittle and would cause the decrease in the elongation at break of blends. However, the elongation at break was improved by the addition of MAH grafted onto EPDM because of the improved morphology of EPDM phase. This would stabilize and improve the cohesion between phases.

Effect of blending methods (one-step blending and two-step blending), on the elongation at break of PET/EPDM blends is presented in Table 4.11. Normally, MAH could cause the scission of PET chain so less amount of MAH is remained to graft onto EPDM. Consequently, the one-step blending gave the lower elongation than two-step blending.

Table 4.10 Elongation of PET/EPDM-g-MAH blends at various MAH contents for PET/EPDM ratio of 80/20.

Ratio	Elongation of break (%)	
	PET/Buna6740	PET/Buna3950
100/0	2.33	2.33
80/20	1.23	1.62
80/20-g-1*	1.31	2.16
80/20-g-3*	2.06	2.18
80/20-g-5*	2.99	2.58
80/20/3**	1.44	2.13

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

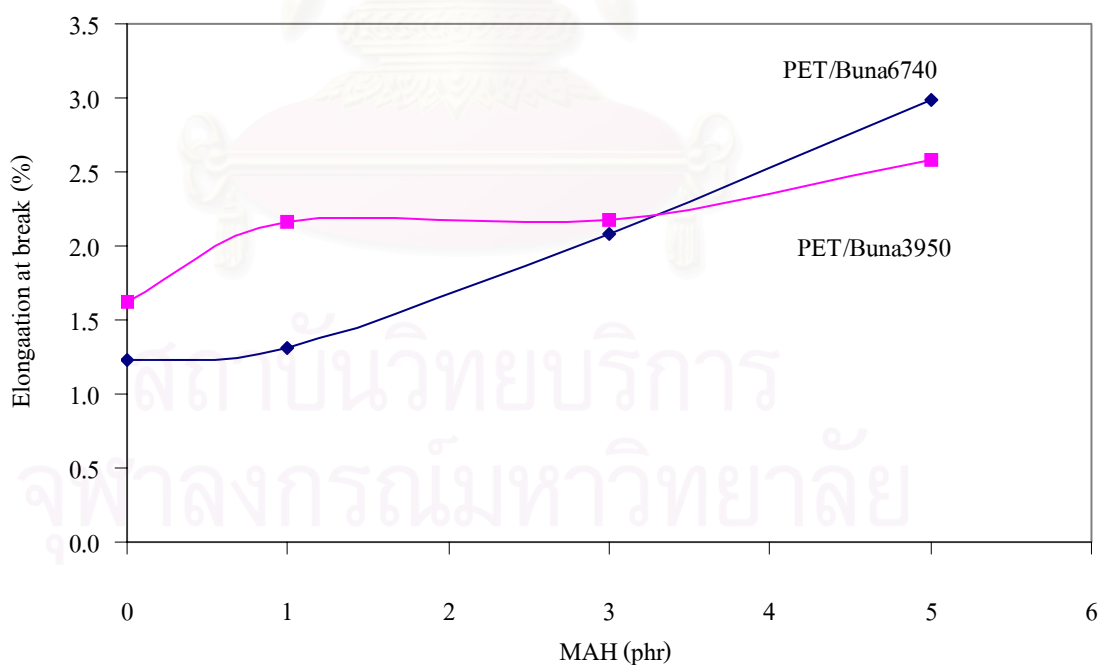


Figure 4.5 Effect of MAH content on elongation at break of PET/EDPM-g-MAH blends.

Table 4.11 Elongation of PETBbuna3950-g-MAH blends at 3 phr MAH and various PET/EPDM blend ratios.

	Elongation of break	
	(%)	
	PET/Buna3950-g-MAH at 80/20	PET/Buna3950-g-MAH at 70/30
0	1.23	1.43
Buna-g-MAH 3 phr (Two-step blending)	2.06	2.23
MAH 3 phr (One-step blending)	1.44	1.35

4.4 Miscibility Study of PET/EPDM-g-MAH Blends by DSC Measurement

Effect of MAH content on glass transition temperature (T_g) and melting temperature (T_m) of PET/EPDM-g-MAH was investigated. The values of T_g and T_m of PET/Buna3950-g-MAH and PET/Buna6740-g-MAH are presented in Tables 4.12 and 4.13.

Figures 4.6, 4.7 and Tables 4.12 and 4.13 show T_g and T_m of PET/Buna3950-g-MAH blends and of PET/Buna6740-g-MAH blends with variation of MAH content from 0 to 5 phr. In case of blend without MAH, average glass transition temperature of the blend was not different from original PET. T_g of the blend with addition of MAH content at 1-5 phr in EPDM does not significant change from original PET. This may be explained that EPDM-g-MAH had little improvement in the miscibility of EPDM in amorphous region of PET. PET is the semicrystalline polymer with melting temperature of 252.7°C. No marked change was observed for the melting point of two phases in these immiscible and miscible blends. From Figure 4.8, the T_g and T_m of PET/Buna3950 at 80/20 blend ratio with 3 phr MAH for different blending method were compared. Blends from two-step blending and one-step blending exhibited no significant change in T_g from original PET. For T_m of the blends, no change was observed in the blends.

The effect of MAH contents on the miscibility of PET/Buna6740-g-MAH is shown in Table 4.13 and Figure 4.7. T_g and T_m of the blends does not change significantly from original PET when MAH content increased from 0 to 5 phr. For the two different blending methods (see Figure 4.9), change of T_g and T_m of the blends was not observed. Therefore, it could not be concluded about PET/EPDM-g-MAH blend miscibility by DSC measurement. Eventhough, the miscibility in SEM study could be observed (Figures 4.10-4.21).

Table 4.12 T_g and T_m of PET/Buna3950-g-MAH blends at various MAH contents.

PET/Buna3950		
Ratio	T_g (°C)	T_m (°C)
PET	73.5	252.7
Buna3950	-	38.5
80/20	72.3	254.7
80/20-g-1*	71.8	253.8
80/20-g-3*	71.2	254.5
80/20-g-5*	71.3	254.6
80/20/3**	71.6	253.5

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

Table 4.13 T_g and T_m of PET/Buna6740-g-MAH blends at various MAH contents

PET/Buna6740		
Ratio	T_g (°C)	T_m (°C)
PET	73.5	252.7
Buna6740	-	40.5
80/20	72.5	253.7
80/20-g-1	71.7	253.7
80/20-g-3	71.6	254.8
80/20-g-5	71.5	254.7
80/20/3	71.7	255.7

* Buna-g-MAH, 1,3,5 phr MAH (Two-step blending).

** 3 phr MAH (One-step blending).

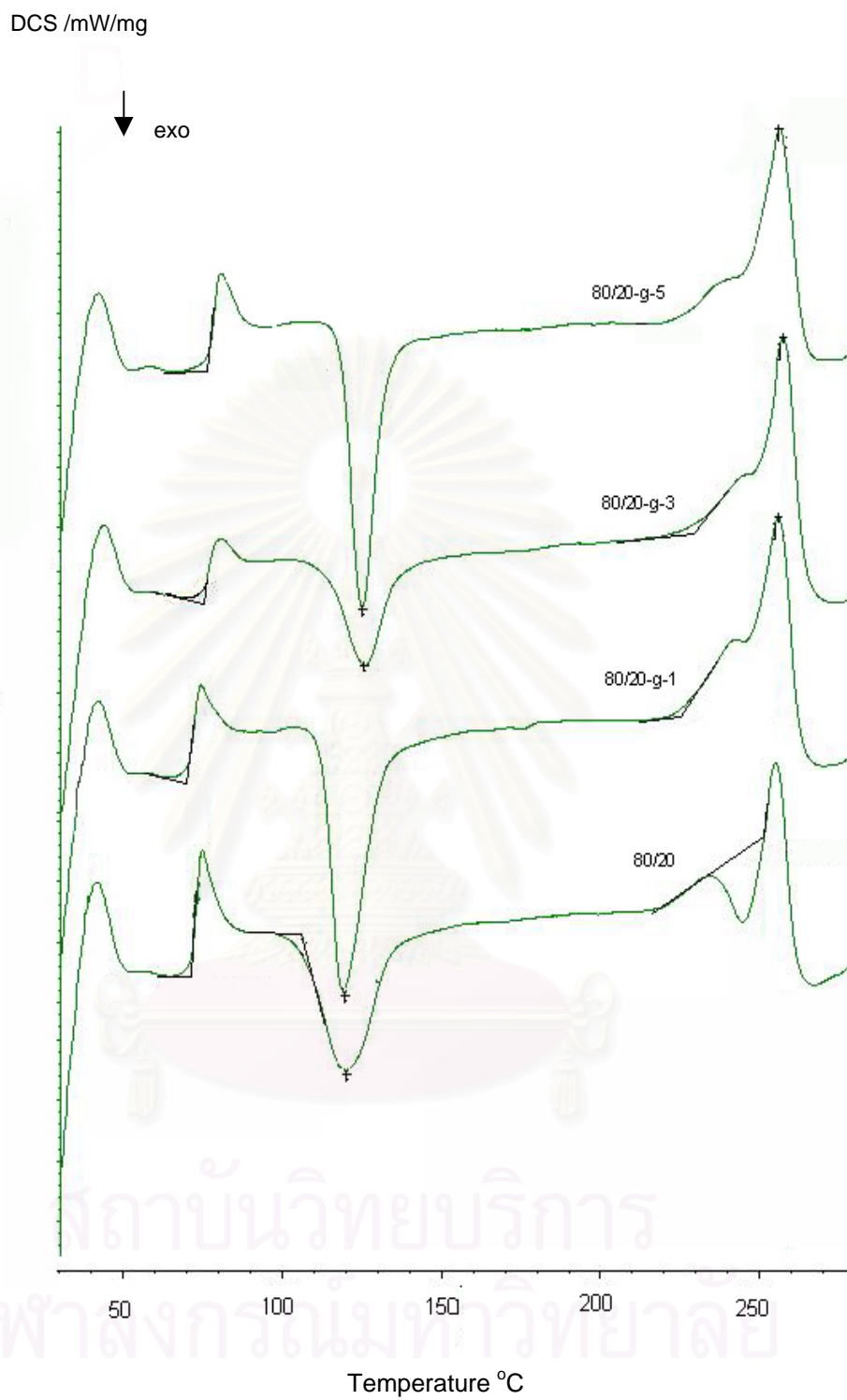


Figure 4.6 DSC thermograms of PET/ Buna3950-g-MAH blends at various MAH contents.

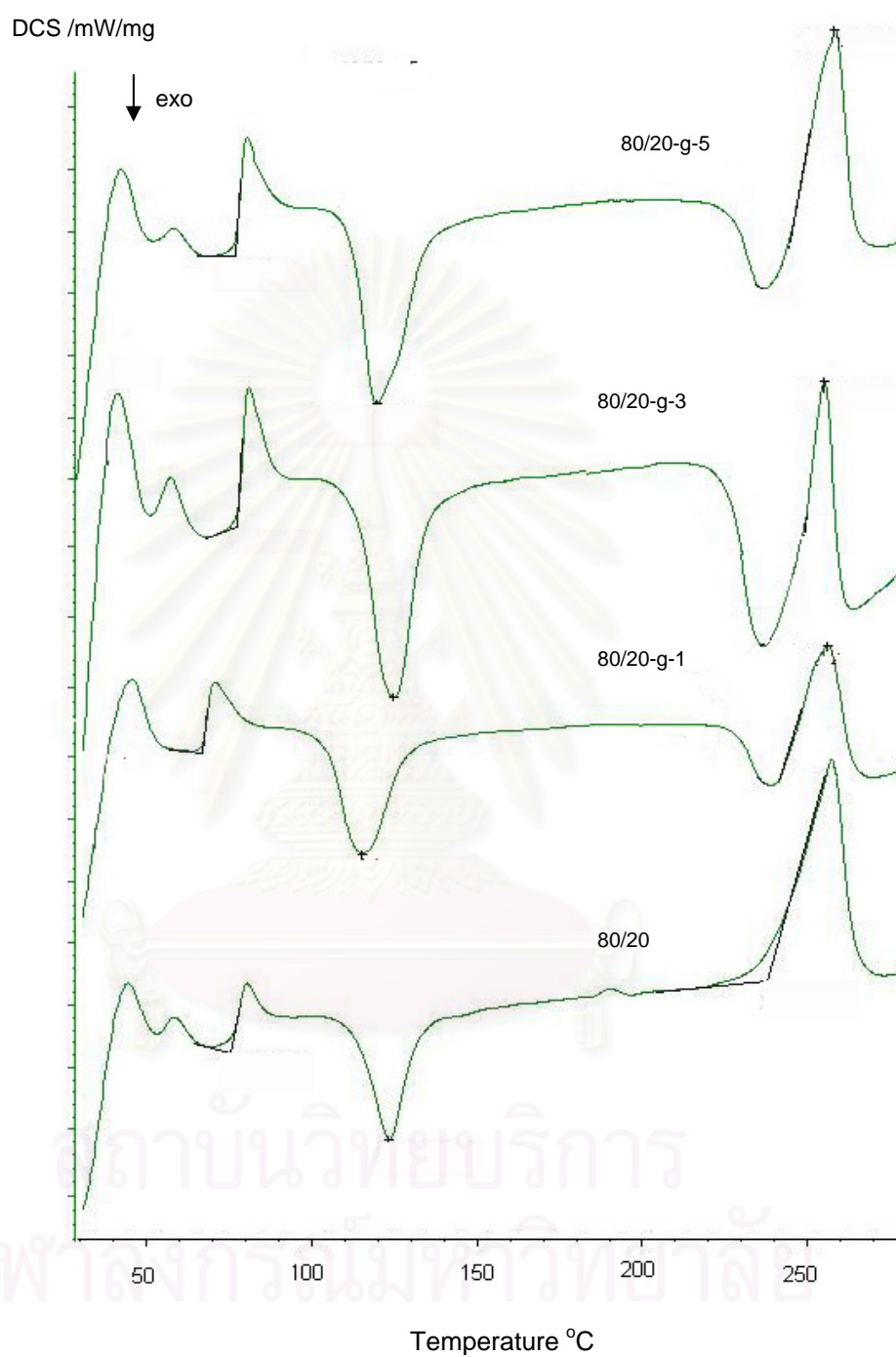


Figure 4.7 DSC thermograms of PET/ Buna6740-g-MAH blends at various MAH contents.

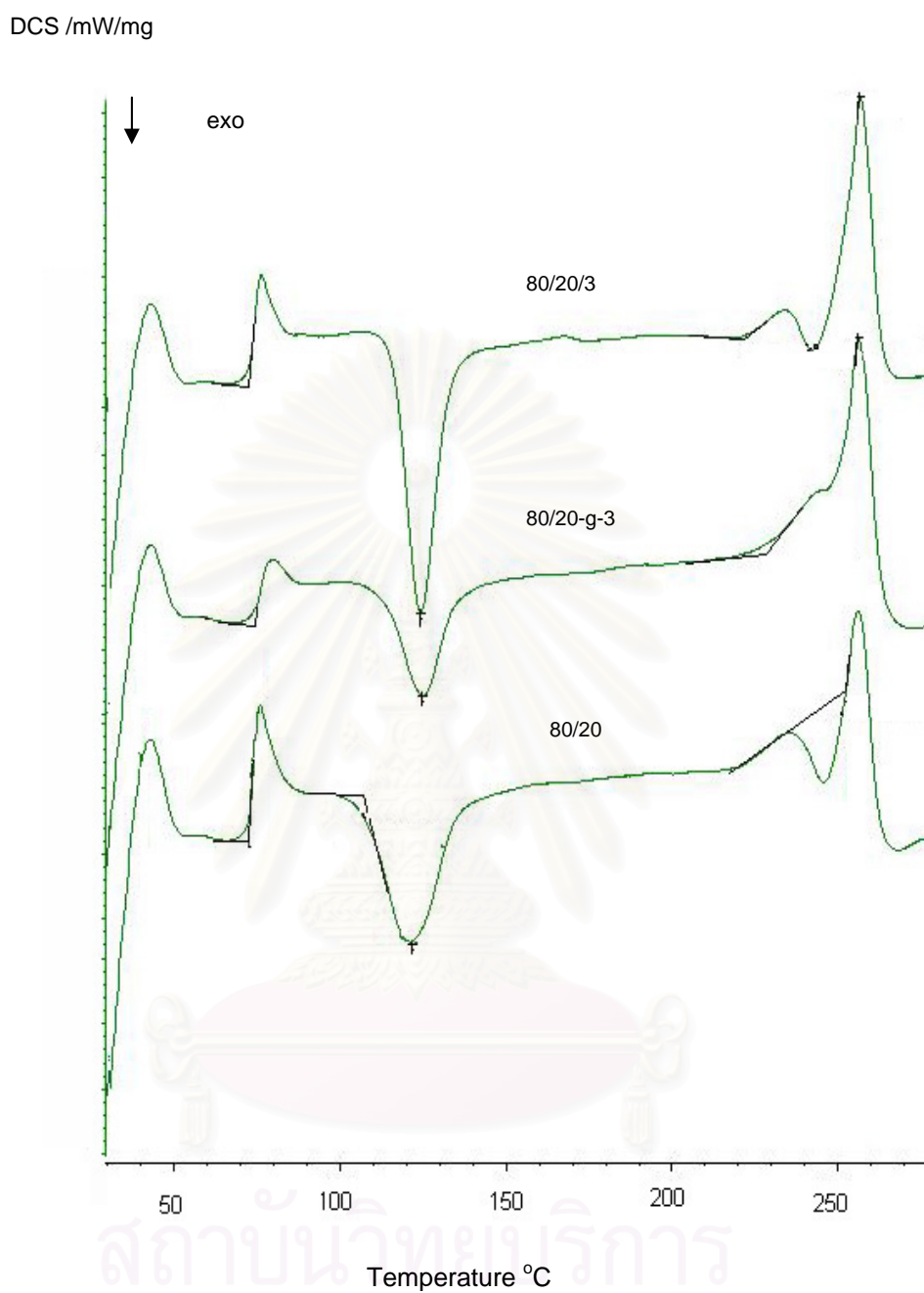


Figure 4.8 DSC thermograms of PET/ Buna3950-g-MAH blends for various blending methods.

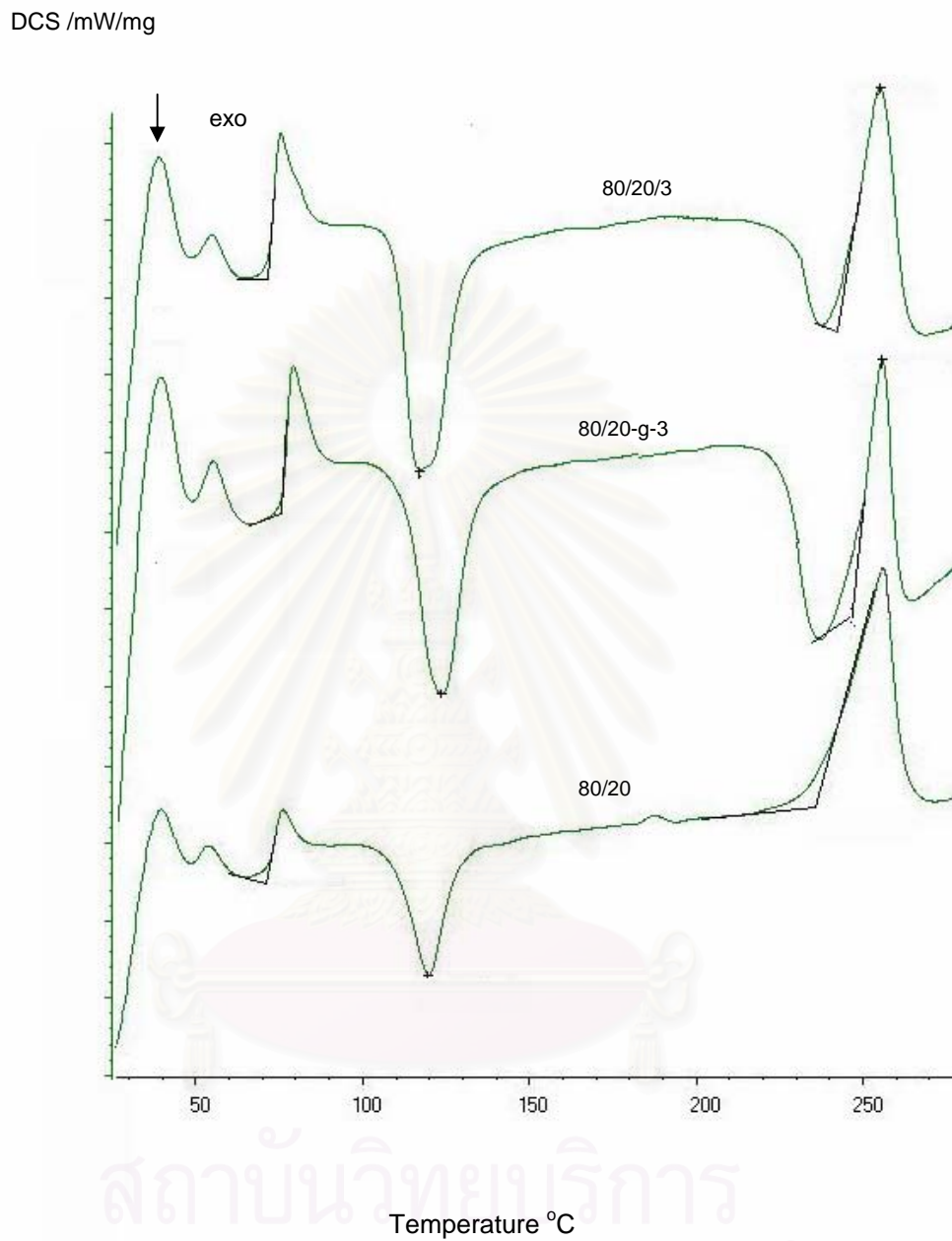


Figure 4.9 DSC thermograms of PET/ Buna6740-g-MAH blends for various blending methods.

4.5 Morphology of PET/EPDM-g-MAH Blend

The scanning electron microscope was employed to investigate morphology and compatibility of PET/ EPDM-g-MAH blends. Figures 4.10-4.21 show the surface of PET/ EPDM-g-MAH blends at MAH content of 0-5 phr.

In SEM micrograph of 80/20 PET/Buna3950 blend without MAH grafted onto EPDM (Figure 4.10), there exists a sharp interface between dispersed EPDM and PET matrix. Dispersed EPDM had an elongated shape and very broad size distribution. An elongated shape of EPDM might be formed by coalescence of several EPDM. Moreover, there are many voids from which the dispersed EPDM was pulled out. This is typical morphology of an incompatible blend.

When Buna3950-g-MAH was blended with PET (Figures 4.11-4.14), the dispersed size of EPDM had a spherical shape and the size was significantly reduced. As the content of MAH increased, this adhesion between matrix and dispersed phase seemed to be stronger. The reaction of maleic anhydride group grafted on EPDM and hydroxyl (or carboxyl) group of PET improved the compatibility between PET and EPDM.

As the content of Buna3950 increased to 30-weight fraction (Figure 4.20), it was observed that there was the large elongated EPDM that formed by coalescence of several EPDM droplets. The morphology of 70/30-g-3 PET/Buna3950 (Figure 4.21) changed and the boundary between PET and EPDM phase was not observed.

From SEM micrograph of 80/20 PET/Buna6740 blend without MAH grafted onto EPDM (Figure 4.16), it was observed that there were a large number of elongated EPDM in PET matrix. When Buna6740-g-MAH was blended in PET (Figure 4.16-4.19), the dispersed size of EPDM had spherical shape and reduced with increasing MAH content but was also bigger than that of PET/Buna3950. For two blending methods, the blends of 80/20 PET/Buna3950 and PET/EPDM6740 with 3 phr MAH do not exhibit clear morphology. The effect of the blending method on morphology of blends could not be observed and discussed.

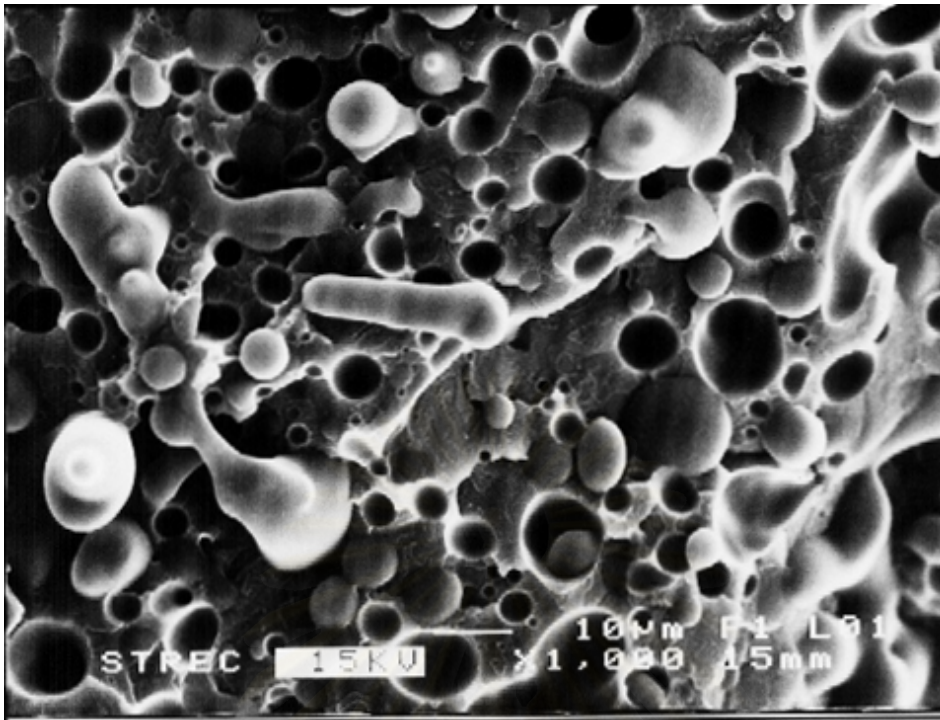


Figure 4.10 Scanning electron micrograph of PET/Buna3950 blend at 80/20.



Figure 4.11 Scanning electron micrograph of PET/Buna3950 blend at 80/20, using 1 phr MAH grafted onto EPDM.

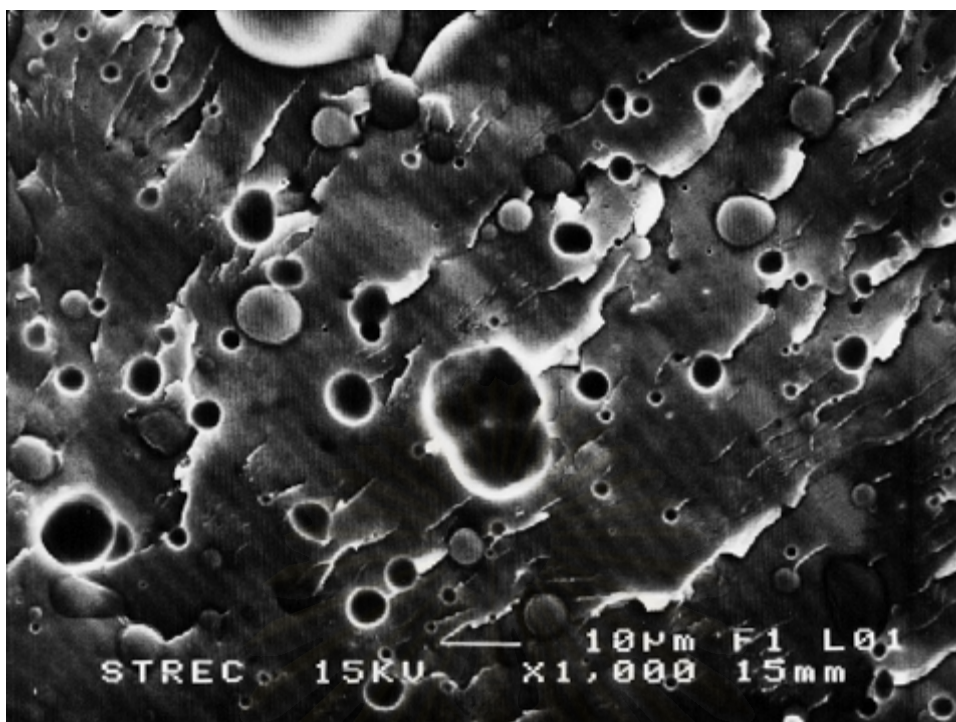


Figure 4.12 Scanning electron micrograph of PET/Buna3950 blend at 80/20, using 3 phr MAH grafted onto EPDM (Two-step blending).

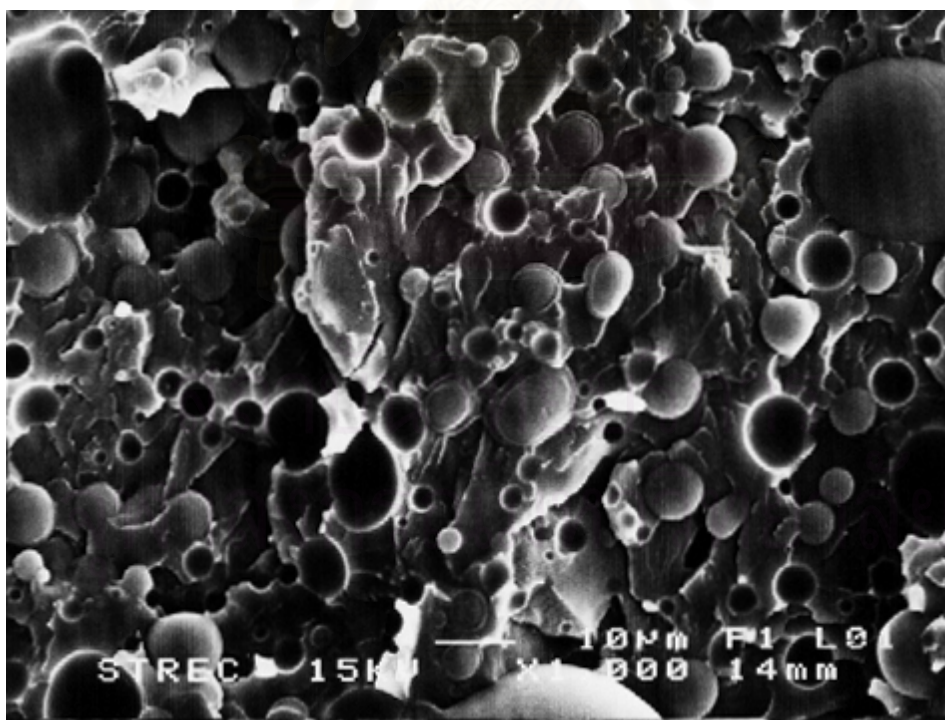


Figure 4.13 Scanning electron micrograph of PET/Buna3950 blend at 80/20, using 3 phr MAH grafted onto PET/EPDM (One-step blending).



Figure 4.14 Scanning electron micrograph of PET/Buna3950 blend at 80/20, using 5 phr MAH grafted onto EPDM.

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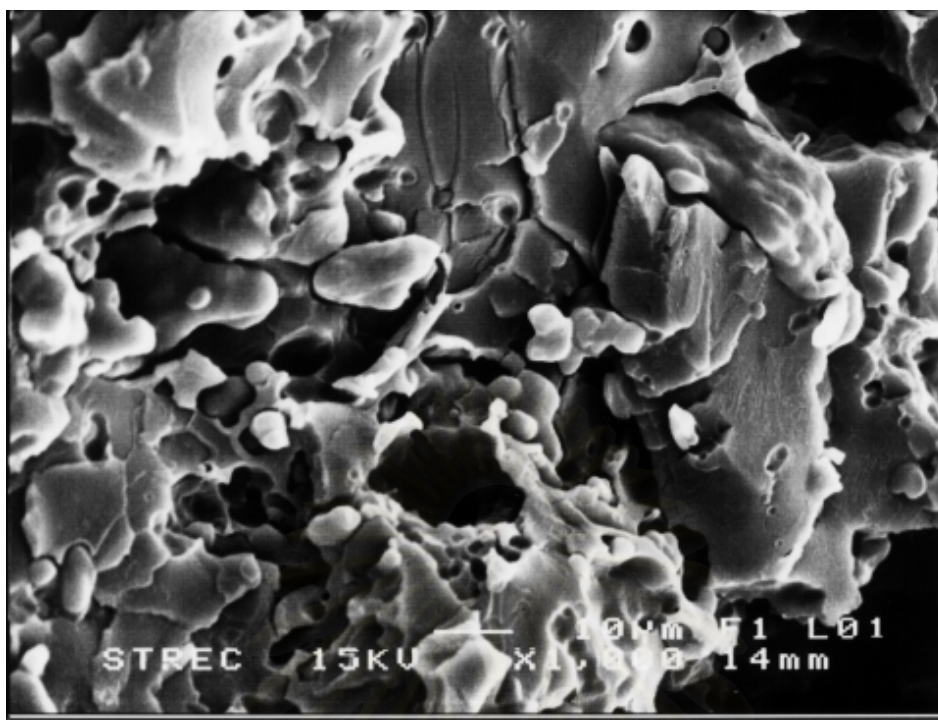


Figure 4.15 Scanning electron micrograph of PET/Buna6740 blend at 80/20.

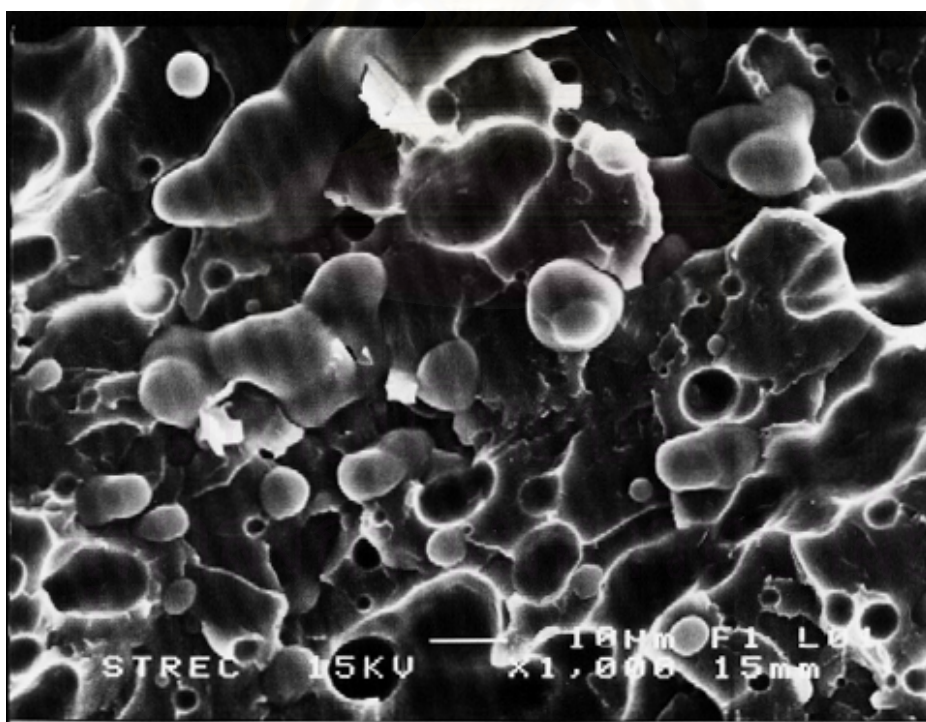


Figure 4.16 Scanning electron micrograph of PET/Buna6740 blend at 80/20, using 1 phr MAH grafted onto EPDM.

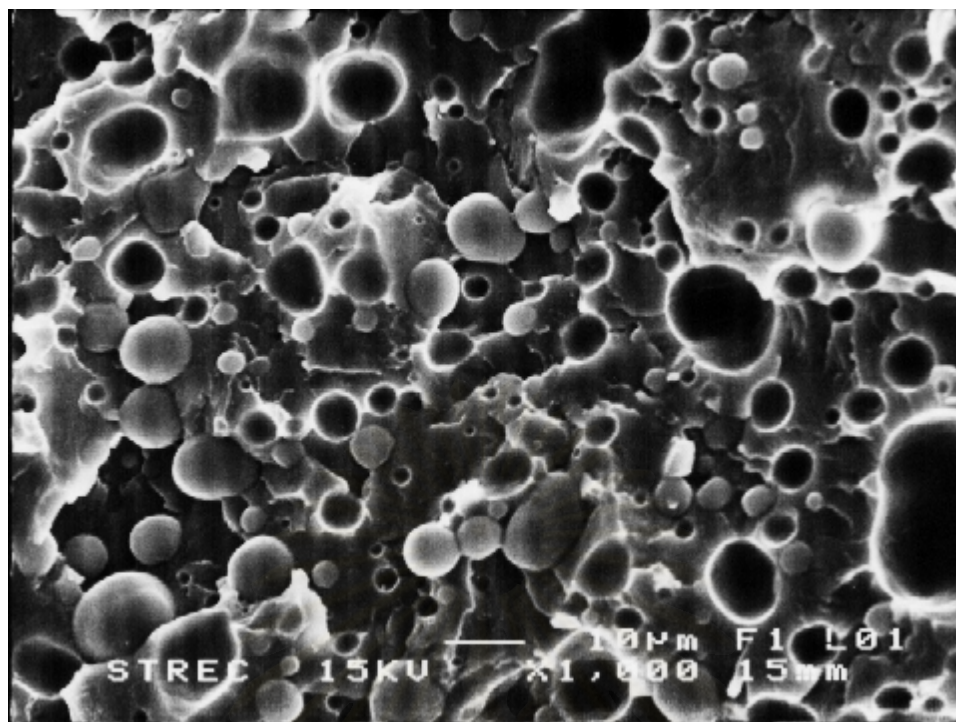


Figure 4.17 Scanning electron micrograph of PET/Buna6740 blend at 80/20, using 3 phr MAH grafted onto EPDM (Two-step blending).

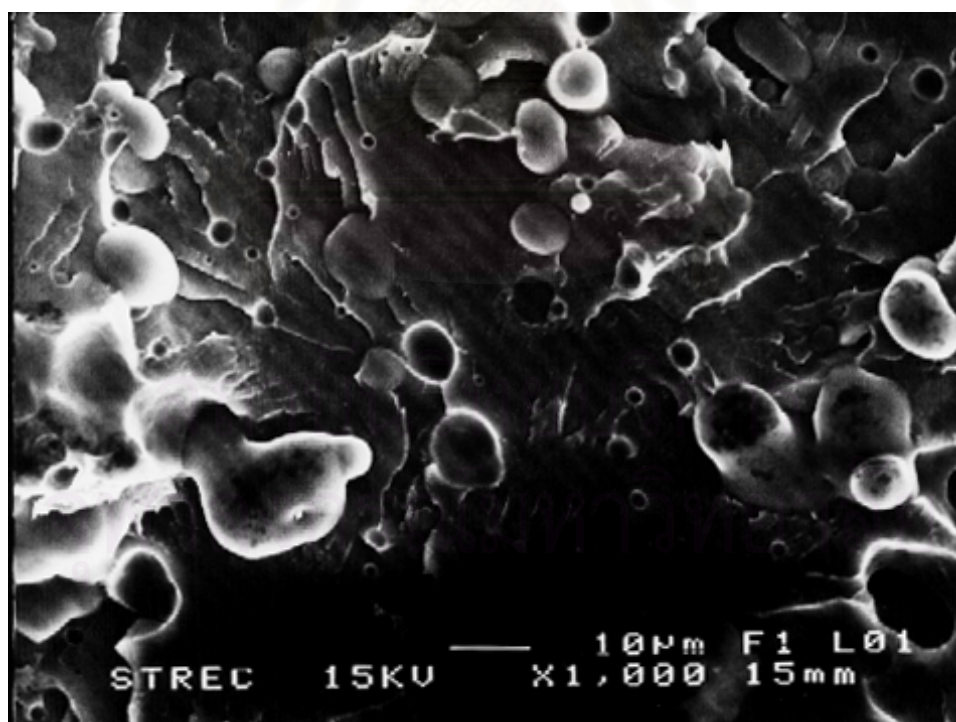


Figure 4.18 Scanning electron micrograph of PET/Buna6740 blend at 80/20, using 3 phr MAH grafted onto PET/EPDM (One-step blending).

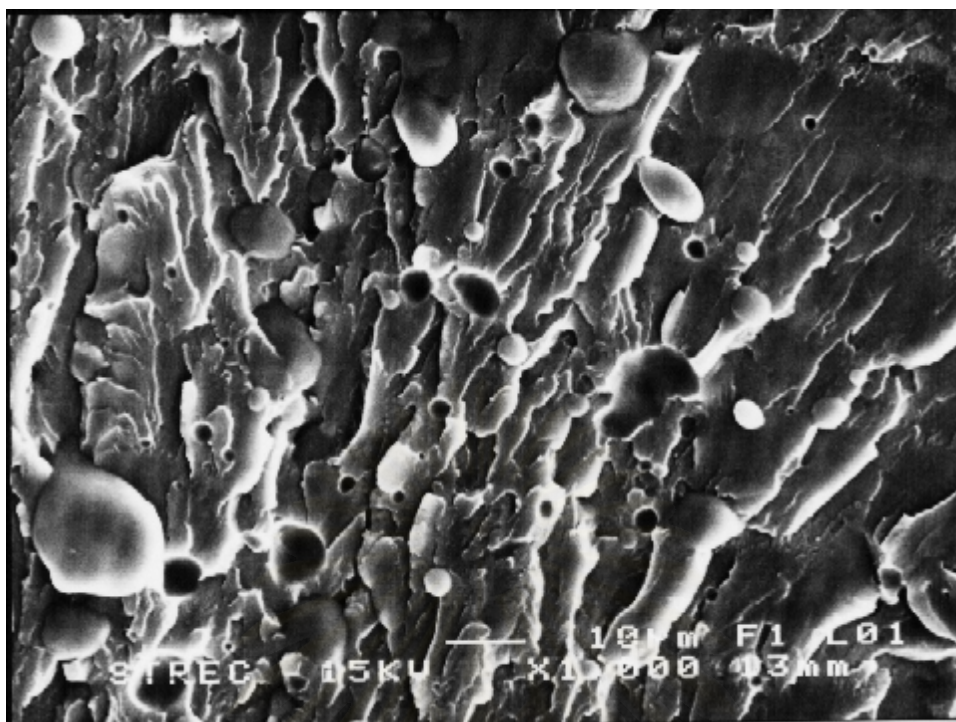


Figure 4.19 Scanning electron micrograph of PET/Buna6740 blend at 80/20, using 5 phr MAH grafted onto EPDM.

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Figure 4.20 Scanning electron micrograph of PET/Buna3950 blend at 70/30.

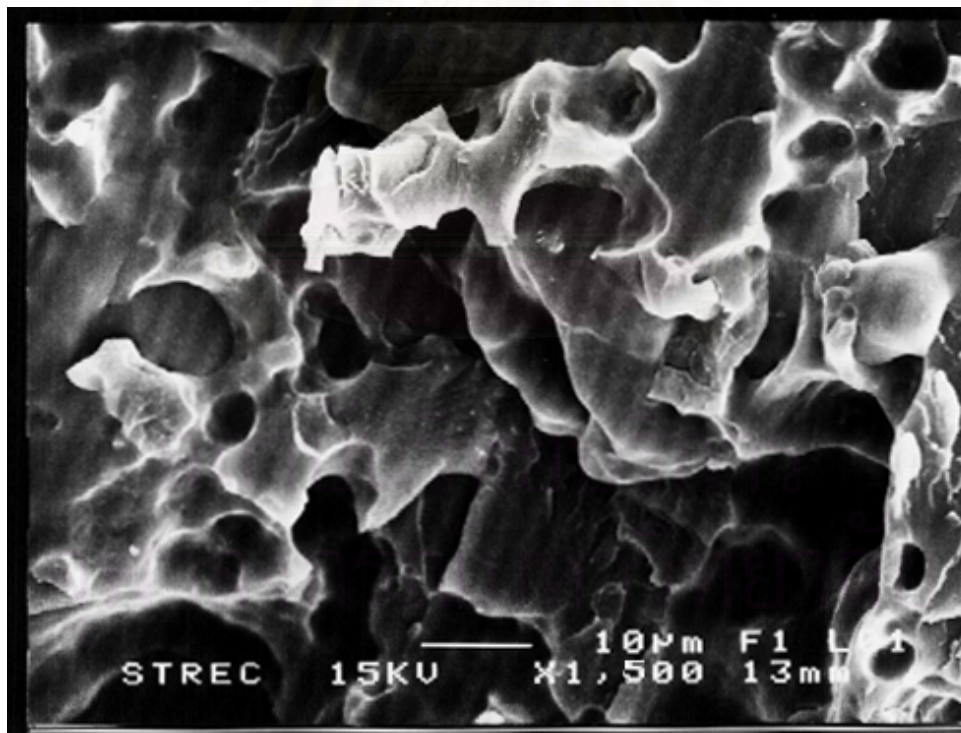


Figure 4.21 Scanning electron micrograph of PET/Buna3950 blend at 70/30, using 3 phr MAH grafted onto EPDM.

4.6 Recycled PET/EPDM-g-MAH

4.6.1 Melt Flow Index of Recycled PET/EPDM-g-MAH

The melt flow index of recycled PET/EPDM-g-MAH is presented in Table 4.14. In general, the melt flow index of recycled PET is very high because of the chain scission or degradation that occurred during the past processing.

The improvement of melt flow index was made by using EPDM-g-MAH in blends at PET/EPDM of blend ratio 70/30 and 80/20. The EPDM-g-MAH improved melt flow index of recycled PET in similar manner as origin PET as discussed earlier in section 4.2. MAH in EPDM-g-MAH structure could react with functional groups of PET and EPDM in EPDM-g-MAH entangled in EPDM phase, this leads to reduction of melt flow index in the blends.

Table 4.14 Melt flow index of recycled PET/EPDM-g-MAH blends.

Ratio	Melt Flow index (g/10min)
100/0	67.6
80/20-g-3	59.8
70/30-g-3	43.7

4.6.2 Mechanical Properties of PET/EPDM-g-MAH blend

Mechanical properties of PET/EPDM-g-MAH are shown in Table 4.15. For the recycled PET/EDPM-g-MAH at ratio 70/30-g-3 and 80/20-g-3, the improvement of impact strength was made. The impact strength increased with increasing EPDM-g-MAH content, because the elastomer is significant impact modifier in the blends.

The tensile strength of recycled PET/EPDM-g-MAH was lower than that of recycled PET because the property of elastomer has low tensile strength. When EPDM was blended with PET, the tensile strength of the blend decreased. The elongation at break of recycled PET/EPDM-g-MAH was lower than that of the recycled PET. The explanation is similar to that for blends of virgin PET which already discussed earlier in section 4.3.

Table 4.15 Mechanical properties of recycled PET/EPDM-g-MAH blends.

Ratio	Impact strength (J/m)	Tensile strength (MPa)	Elongation at break (%)
100/0	8.72	14.85	1.75
80/20-g-3	10.54	3.30	1.65
70/30-g-3	24.00	4.47	1.38

CHAPTER V

CONCLUSION AND SUGGESTION FOR FURTHER WORK

Polyethylene terephthalate (PET) is semicrystalline thermoplastic polyester having high notch sensitivity. The effective way to enhance the impact strength is provided by the dispersion of rubber phase within the PET matrix. In order to obtain fine particle size of dispersed phase, the use of reactive compatibilization was found to be essential. The compatibilization efficiency of two functional groups (maleic anhydride, MAH) regarding to the morphology refinement of both basic PET/EPDM blends (Buna3950 and Buna6740) were evaluated. EPDM containing MAH group was blended with PET. In view of the blend morphology, dispersed particles of the PET/EPDM-g-MAH were finer than those of PET/EPDM blends, and the PET/EPDM-g-MAH blend showed homogeneous dispersions and even partial adhesions between the dispersed and matrix phases. The compatibilizer (EPDM-g-MAH) could reduce the interfacial tension in the melt, causing an emulsifying effect and leading to extremely fine dispersion of one phase in another. Another effect is to stabilize the dispersed phase during annealing, again by modifying the phase-boundary interface.

From thermal analysis, the compatibilized blend could not be confirmed because T_g and T_m of the blends did not significantly shift from original PET. In melt flow index measurements, the decrease in of melt flow index was observed for the PET/EPDM-g-MAH blends, which were also related to the chemical reaction. In mechanical properties, the PET/EPDM-g-MAH blends showed the improved notch-Izod impact strength over PET and PET/EPDM blends. Optimum impact strength of the blends was obtained when the MAH content in the grafting reaction was 3 phr.

Tensile strength and elongation at break of the PET/EPDM-g-MAH blends are functions of the dispersed phase size. They depend solely on MAH content in EPDM

elastomer in the blends. EPDM-g-MAH effect is to increase the adhesion at phase boundaries, giving improved stress transfer of blends.

The blending methods at 3 phr MAH, two-step blending gave higher mechanical strength and melt flow index than that one-step blending. One-step blending caused only small amount of MAH diffused to react with functional groups of PET.

Suggestion for Further Work

1. In this study, the optimum MAH content was 3 phr when the diene content of EPDM was 11.4 %. The % MAH grafted onto EPDM was about 1.83 %. The grafting MAH onto EPDM produced via single screw extruder, another processing methods is suggested to increase the grafting degree of EPDM-g-MAH.

2. In this study, the blend miscibility was limited by blending technique, the other blending technique, twin screw extruder is suggested to improve miscibility of blends.

3. The other elastomer can be used to improve the mechanical strength of PET: e.g. nitrile rubber (NBR) or styrene-butadiene-rubber (SBR). The properties of PET blends will be improved for using in a wide range application.

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APPENDICES

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APPENDIX A

Statistic of Data

Standard deviation (s), Equation (A1) and t distribution was to determine the distribution of experiments values [29].

$$S = \frac{\sum \sqrt{(X_i - X)^2}}{n-1} \quad (A1)$$

where n is the number of sample,

X_i is the experimental of I experiment,

X is the arithmetic experimental mean of a set of values.

The true data (μ) was calculated following the equation (A2). The t-values were obtain from the tabulated statistical table.

$$\mu = X \pm \frac{(tSD)}{\sqrt{n}} \quad (A2)$$

From the statistical table:

At 90% confidence, n=3, to obtain t = 2.92.

At 90% confidence, n=5, to obtain t = 2.13.

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APPENDIX B
Maleic Anhydride Grafting

Table B1. Maleic Anhydride Grafting on EPDM (Buna3950), using KOH 0.071 N

MAH on EPDM	Weight EDPM-g-MAH	Organic phase			Solution phase		Gel phase
		KOH titrated in Toluene	% MAH grafted onto EPDM	% MAH reacts	KOH titrated in solution	% MAH unreacts	% crosslinking
1phr	1.05	1.25	0.83	43.67	0.55	19.22	37.11
	1.03	1.30	0.88	45.42	0.50	17.47	37.11
3 phr	1.08	2.85	1.84	33.19	0.35	12.23	54.58
	1.01	2.50	1.73	29.11	0.30	10.48	60.40
5 phr	1.08	2.90	1.88	20.26	0.15	5.24	74.50
	1.00	2.55	1.78	17.82	0.20	6.99	75.19

Table B2. Maleic Anhydride Grafting on EPDM (Buna6740), using KOH 0.10 N

MAH on EPDM	weight EDPM-g-MAH	Organic phase			Solution phase		Gel phase
		KOH titrated in Toluene	% MAH grafted onto EPDM	% MAH reacts	KOH titrated in solution	% MAH unreacts	% crosslinking
1phr	1.01	0.70	0.70	35.66	0.50	25.47	38.87
	1.06	0.80	0.77	40.75	0.45	22.92	36.33
3 phr	1.01	1.10	1.12	18.68	0.65	11.04	70.29
	1.03	1.10	1.09	18.68	0.80	13.58	67.74
5 phr	1.01	1.11	1.12	11.31	0.18	1.83	86.86
	1.02	1.10	1.10	11.21	0.15	1.53	87.27

APPENDIX C

Melt Flow index

Table C1. Melt flow index of PET/Buna3950-g-MAH at ratio 80/20

	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	57.11	47.38	49.78	50.75	41.77	57.79
2	64.37	63.3	44.70	42.96	41.32	51.74
3	69.02	52.69	54.41	50.83	47.69	63.01
4	70.70	52.60	51.31	43.72	45.90	41.60
5	54.34	49.98	42.91	39.6	44.39	51.19
Average	63.11	53.19	48.62	45.57	44.21	53.07

Table C2. Melt flow index of PET/Buna3950-g-MAH at ratio 70/30

	100/0	70/30	70/30-g-3	70/30/3
1	57.11	56.63	33.84	47.71
2	64.37	49.58	43.43	45.49
3	69.02	44.28	42.86	39.94
4	70.70	42.77	42.13	40.43
5	54.34	60.79	39.02	52.60
Average	63.11	50.81	40.26	45.23

Table C3. Melt flow index of PET/Buna6740-g-MAH at ratio 80/20

	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	57.11	62.83	53.62	65.54	45.37	63.76
2	64.37	60.35	58.08	50.00	54.25	59.06
3	69.02	59.04	60.48	58.24	62.76	69.04
4	70.70	60.20	61.22	54.70	65.42	55.02
5	54.34	61.09	67.69	66.90	66.89	55.99
Average	63.11	60.7	60.22	59.80	58.94	60.57

Table C4. Melt flow index of recycled PET/Buna3950-g-MAH

	100/0	70/30-g-3	80/20-g-3
1	71.18	39.49	56.18
2	60.23	41.42	57.13
3	64.55	42.88	57.70
4	70.99	44.86	61.64
5	71.18	50.10	66.48
Average	67.63	43.75	59.83

APPENDIX D

Izod Impact strength

Table D1. Izod Impact strength of PET/Buna3950-g-MAH at ratio 80/20

No.	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	11.42	14.545	11.19	14.28	13.53	8.89
2	11.85	11.189	12.03	15.03	10.00	8.89
3	11.85	11.852	12.31	17.77	12.12	10.49
4	12.14	11.94	12.31	17.91	12.03	11.59
5	12.50	10.769	12.31	19.58	12.31	11.85
Average	11.96	11.99	12.03	16.918	12.00	10.34
SD	0.41	1.49	0.48	2.20	1.27	1.42

Table D2. Izod Impact strength of PET/Buna3950-g-MAH at ratio 70/30

No.	100/0	70/30	70/30-g-3	70/30/3
1	11.42	11.59	29.63	21.21
2	11.85	12.12	29.63	21.33
3	11.85	14.60	30.08	23.70
4	12.14	14.82	32.84	24.24
5	12.50	14.82	33.08	33.07
Average	11.96	13.59	31.05	24.71
SD	0.41	1.59	1.75	4.87

Table D3. Izod Impact strength of PET/Buna6740-g-MAH at ratio 80/20

No.	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	11.42	11.59	11.85	11.85	12.301	12.06
2	11.85	11.59	12.22	13.99	14.71	13.14
3	11.85	12.03	14.29	14.49	14.07	12.77
4	12.14	14.82	14.49	15.04	11.43	13.24
5	12.50	11.43	14.71	17.14	14.82	11.27
Average	11.96	12.29	13.51	14.50	13.47	12.48
SD	0.41	4.13	1.36	1.91	1.52	0.83

Table D4. Izod Impact strength of recycled PET/Buna3950-g-MAH

No.	100/0	70/30-g-3	80/20-g-3
1	8.57	20.00	11.05
2	8.57	22.86	10.74
3	8.70	25.72	10.74
4	8.76	25.72	10.44
5	9.02	25.72	9.72
Average	8.72	24.00	10.54
SD	0.19	2.56	0.45

APENDIX E

Tensile Strength

Table E1. Tensile Strength of PET/Buna3950-g-MAH at ratio 80/20

No.	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	24.60	6.67	9.69	12.51	15.38	11.51
2	18.17	8.05	9.30	12.22	15.78	11.21
3	25.26	10.21	10.58	13.46	15.43	12.58
4	28.11	9.67	12.54	14.32	15.80	13.57
5	26.11	11.00	15.12	11.78	13.73	12.58
Average	24.45	9.16	11.44	12.86	15.22	12.29
SD	3.75	1.68	2.41	1.02	0.86	0.95

Table E2. Tensile Strength of PET/Buna3950-g-MAH at ratio 70/30

No.	100/0	70/30	70/30-g-3	70/30/3
1	24.60	6.62	6.31	5.75
2	18.17	6.24	5.42	5.99
3	25.26	6.53	7.22	5.26
4	28.11	5.51	5.79	4.88
5	26.11	6.86	5.56	6.01
Average	24.45	6.28	6.06	5.58
SD	3.75	0.50	0.73	0.49

Table E3. Tensile Strength of PET/Buna6740-g-MAH at ratio 80/20

No.	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	24.60	9.65	11.01	16.70	14.18	7.16
2	18.17	10.00	12.09	12.79	16.01	7.69
3	25.26	9.80	11.86	12.36	15.06	6.84
4	28.11	8.21	9.58	15.68	15.36	7.79
5	26.11	8.38	13.23	12.75	15.13	7.79
Average	24.45	9.21	11.55	14.06	15.15	7.45
SD	3.75	0.85	1.35	1.98	0.66	0.43

Table E4. Tensile Strength of recycled PET/Buna3950-g-MAH

No.	100/0	70/30-g-3	80/20-g-3
1	15.74	2.28	2.70
2	12.83	4.03	2.83
3	17.90	3.39	5.62
4	12.94	3.49	5.93
5			5.28
Average	14.85	3.30	4.47
SD	2.43	0.73	1.58

APENDIX F

Elongation at Break

Table F1. Elongation at Break of PET/Buna3950-g-MAH at ratio 80/20

No.	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	2.27	1.19	2.06	1.72	2.52	2.51
2	1.83	1.29	1.77	2.11	2.84	1.80
3	2.34	2.12	2.57	2.62	2.77	2.26
4	2.71	1.70	2.96	1.82	2.53	2.04
5	2.50	1.80	1.44	2.61	2.26	2.03
Average	2.33	1.62	2.16	2.18	2.58	2.13
SD	0.32	0.38	0.61	0.43	0.23	0.27

Table F2. Elongation at Break of PET/Buna3950-g-MAH at ratio 70/30

No.	100/0	70/30	70/30-g-3	70/30/3
1	2.27	1.08	2.57	1.31
2	1.83	1.09	2.23	1.39
3	2.34	1.38	2.84	1.40
4	2.71	1.09	1.98	1.42
5	2.50	2.50	1.51	1.24
Average	2.33	1.43	2.23	1.35
SD	0.32	0.61	0.52	0.07

Table F3. Elongation at Break of PET/Buna6740-g-MAH at ratio 80/20

No.	100/0	80/20	80/20-g-1	80/20-g-3	80/20-g-5	80/20/3
1	2.27	1.29	1.86	2.24	2.42	1.18
2	1.83	1.40	1.24	1.76	3.89	1.35
3	2.34	1.34	1.97	1.91	2.88	1.43
4	2.71	1.03	4.95	2.68	2.99	1.90
5	2.50	1.10	1.54	1.83	2.77	1.34
Average	2.33	1.23	1.71	2.09	3.00	1.44
SD	0.32	0.16	0.32	0.37	0.55	0.27

Table F4. Elongation at Break of recycled PET/Buna3950-g-MAH

No.	100/0	70/30-g-3	80/20-g-3
1	1.61	1.62	0.75
2	1.8	2.69	1.17
3	2.17	1.12	1.50
4	1.40	1.17	1.18
5			1.61
Average	1.745	1.65	1.37
SD	0.32	0.73	0.37

VITA

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