

## CHAPTER IV

### RESULTS AND DISCUSSION

Most post-consumer waste consists of mixtures of different polymers. The main parts of post-consumer waste are LDPE and HDPE because these polyolefins are widely used for producing package materials. Although they represent the same polymer class, they are incompatible. Therefore, the properties of a recycled HDPE/LDPE blend do not reach the properties of a single polymer. The situation is further complicated because both polymers have different degrees of degradation, also affecting their compatibility and properties.

The recycled polyethylene (HDPE/LDPE) resins were obtained from various sources with different service histories. In this study, the statistical data analyses were carried out using a personal computer program STAGRAPHICS with one-way analysis of variance (ANOVA) at the 95% confidence level. The results of variance analyses of MFI and tensile properties are shown in Tables 4.1-4.2.

**Table 4.1.** Analysis of Variance of Melt Flow Index at 2.16- and 5-kg Load.

| Source                             | F-ratio | p-value |
|------------------------------------|---------|---------|
| MFI 2.16 vs Vigin HDPE             | 5.26    | 0.0016  |
| MFI 2.16 vs Recycled HDPE (film)   | 0.02    | 0.9844  |
| MFI 2.16 vs Recycled HDPE (bottle) | 12.70   | 0.0002  |
| MFI 2.16 vs Recycled LDPE          | 7.54    | 0.0029  |
| MFI 5 vs Vigin HDPE                | 5.55    | 0.0012  |
| MFI 5 vs Recycled HDPE (film)      | 0.00    | 0.9962  |
| MFI 5 vs Recycled HDPE (bottle)    | 15.03   | 0.0001  |
| MFI 5 vs Recycled LDPE             | 7.06    | 0.0039  |

**Table 4.2** Analysis of Variance of Tensile Properties of the Film in MD and TD.

## a) Film Properties (MD).

| Source  | F-ratio | p-value |
|---|---------|---------|
| Elongation at Break vs Virgin HDPE            | 3.62    | 0.0110  |
| Elongation at Break vs Recycled HDPE (film)   | 0.74    | 0.4887  |
| Elongation at Break vs Recycled HDPE (bottle) | 1.01    | 0.3788  |
| Elongation at Break vs Recycled LDPE          | 0.94    | 0.4050  |
| Stress at Break vs Virgin HDPE                | 12.48   | 0.0000  |
| Stress at Break vs Recycled HDPE (film)       | 0.82    | 0.4509  |
| Stress at Break vs Recycled HDPE (bottle)     | 6.32    | 0.0062  |
| Stress at Break vs Recycled LDPE              | 7.34    | 0.0033  |
| Elongation at Yield vs Virgin HDPE            | 1.68    | 0.1715  |
| Elongation at Yield vs Recycled HDPE (film)   | 1.85    | 0.1795  |
| Elongation at Yield vs Recycled HDPE (bottle) | 1.39    | 0.2674  |
| Elongation at Yield vs Recycled LDPE          | 5.05    | 0.0147  |
| Stress at Yield vs Virgin HDPE                | 4.67    | 0.0031  |
| Stress at Yield vs Recycled HDPE (film)       | 0.25    | 0.7816  |
| Stress at Yield vs Recycled HDPE (bottle)     | 0.98    | 0.3913  |
| Stress at Yield vs Recycled LDPE              | 56.08   | 0.0000  |

## b) Film Properties (TD).

| Source  | F-ratio | p-value |
|---|---------|---------|
| Elongation at Break vs Virgin HDPE            | 3.11    | 0.0218  |
| Elongation at Break vs Recycled HDPE (film)   | 1.04    | 0.3675  |
| Elongation at Break vs Recycled HDPE (bottle) | 1.08    | 0.3567  |
| Elongation at Break vs Recycled LDPE          | 10.52   | 0.0005  |
| Stress at Break vs Virgin HDPE                | 4.33    | 0.0046  |
| Stress at Break vs Recycled HDPE (film)       | 0.07    | 0.9287  |
| Stress at Break vs Recycled HDPE (bottle)     | 4.70    | 0.0190  |
| Stress at Break vs Recycled LDPE              | 12.95   | 0.0002  |
| Elongation at Yield vs Virgin HDPE            | 0.64    | 0.7179  |
| Elongation at Yield vs Recycled HDPE (film)   | 2.87    | 0.0762  |
| Elongation at Yield vs Recycled HDPE (bottle) | 0.43    | 0.6574  |
| Elongation at Yield vs Recycled LDPE          | 2.81    | 0.0803  |
| Stress at Yield vs Virgin HDPE                | 4.08    | 0.0062  |
| Stress at Yield vs Recycled HDPE (film)       | 0.45    | 0.6406  |
| Stress at Yield vs Recycled HDPE (bottle)     | 0.11    | 0.8961  |
| Stress at Yield vs Recycled LDPE              | 71.81   | 0.0000  |

Since the recycled HDPE (bottle grade) exerts, without doubt, a significant effect on MFI, the recycled LDPE gives an interesting stress at yield with an extremely high significance (F-ratio). Table 4.2 also indicates that yield stress of MD of the film is much stronger than that of TD, because the F-ratio of TD is higher. Therefore, the recycled LDPE blend was chosen to study for the effect of the recycled resin on mechanical properties, rheological properties, film properties, thermal behavior, and morphological characteristics of the blended polyethylenes.

#### 4.1 Effect of Virgin HDPE and Recycled Polyethylene Compositions on Mechanical Properties.

##### 4.1.1 Effect on Melt Flow Index (MFI).

Melt flow index is the most common method of characterizing the melt flow property of plastics and is the commercially available instrument to distinguish among different grades of plastics of the same type. In the recycling study, the MFI was used to characterize the occurrence of degradation.

**Table 4.3** Effect of Recycled Polyethylene Resins on MFI.

| Material (%) |                      |                        |               | MFI <sub>2.16/190 °C</sub><br>(g/10 min) | MFI <sub>5/190 °C</sub><br>(g/10 min) |
|--------------|----------------------|------------------------|---------------|--|---------------------------------------|
| Virgin HDPE  | Recycled HDPE (film) | Recycled HDPE (bottle) | Recycled LDPE |  |                                       |
| 100          | -                    | -                      | -             | 0.06±0.01                                | 0.27±0.01                             |
| -            | 100                  | -                      | -             | 0.06±0.01                                | 0.14±0.01                             |
| -            | -                    | 100                    | -             | 2.50±0.03                                | 13.22±0.07                            |
| -            | -                    | -                      | 100           | 4.30±0.07                                | 18.48±0.06                            |
| 70           | 10                   | 10                     | 10            | 0.07±0.01                                | 0.39±0.01                             |
| 60           | 10                   | 10                     | 20            | 0.15±0.01                                | 0.55±0.01                             |
| 50           | 10                   | 10                     | 30            | 0.17±0.02                                | 0.82±0.01                             |
| 50           | 20                   | 20                     | 10            | 0.16±0.01                                | 0.60±0.02                             |
| 40           | 20                   | 20                     | 20            | 0.18±0.01                                | 0.87±0.01                             |
| 30           | 20                   | 20                     | 30            | 0.28±0.01                                | 1.34±0.01                             |
| 30           | 30                   | 30                     | 10            | 0.18±0.01                                | 0.95±0.03                             |
| 20           | 30                   | 30                     | 20            | 0.30±0.01                                | 1.40±0.01                             |
| 10           | 30                   | 30                     | 30            | 0.47±0.01                                | 2.07±0.01                             |

In this study, virgin HDPE and recycled polyethylene resins were blended in the single screw extruder. The effect of virgin HDPE and recycled polyethylene resins at various ratios on MFI is presented in Table 4.1. The F ratios of MFI at two levels of weight load are relatively equal at 5.26 and 5.55, respectively, while F ratios of recycled HDPE film are not significant.

From Table 4.3 and Figure 4.1, the results show that MFI increased with blended recycled low-density polyethylene (LDPE) resins. Degradation of the molecules of recycled HDPE and recycled LDPE might occur to result in lower molecular weights (MW) and increased MFI. It was difficult to improve the MFI of the recycled HDPE to match that of the virgin HDPE. Various shear forces take place during the processing of mixed grades of HDPE and LDPE which degrade them differently leading to broader lower molecular weights and higher MFI. The low-molecular weight PE films can cause a decrease in their film strength.

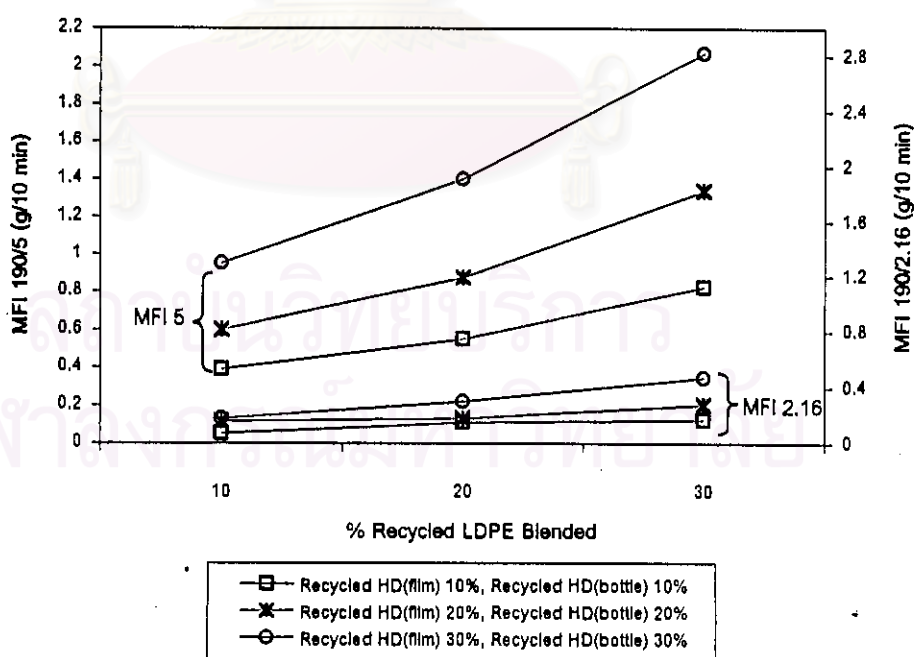


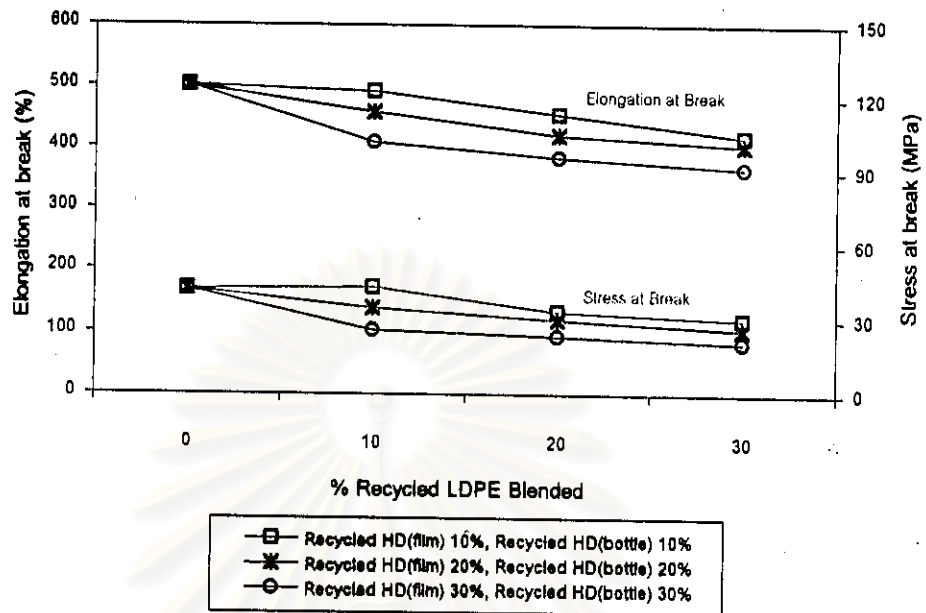
Figure 4.1 Effect of recycled polyethylene resins on MFI.

### 4.1.2 Effect on Tensile Properties.

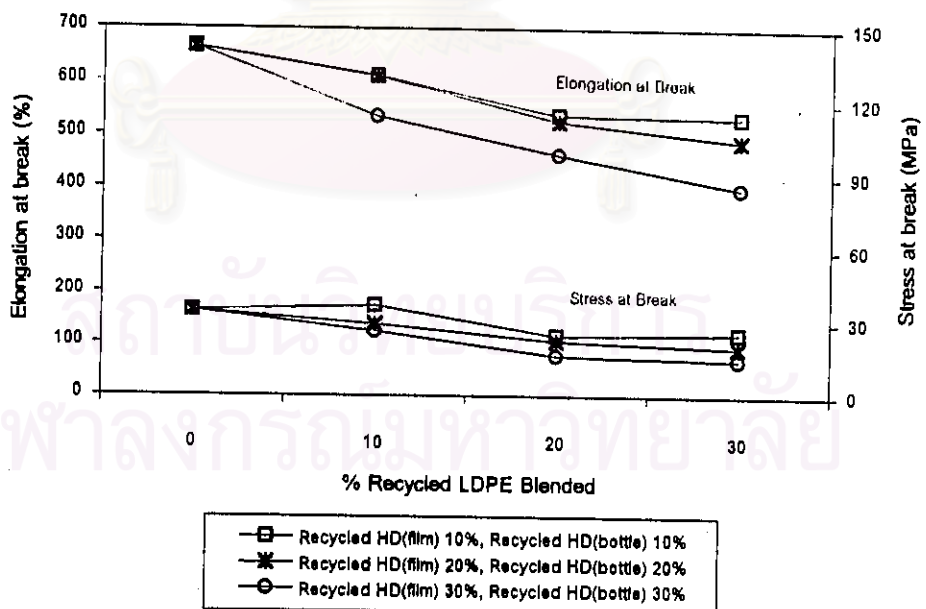
Table 4.4 shows stresses at yield ( $\sigma_y$ ) and at break ( $\sigma_b$ ), elongation at yield ( $\epsilon_y$ ), and elongation at break ( $\epsilon_b$ ) of the blended polyethylene resins.

**Table 4.4** Effect of Recycled Polyethylene Resins on Tensile Properties.

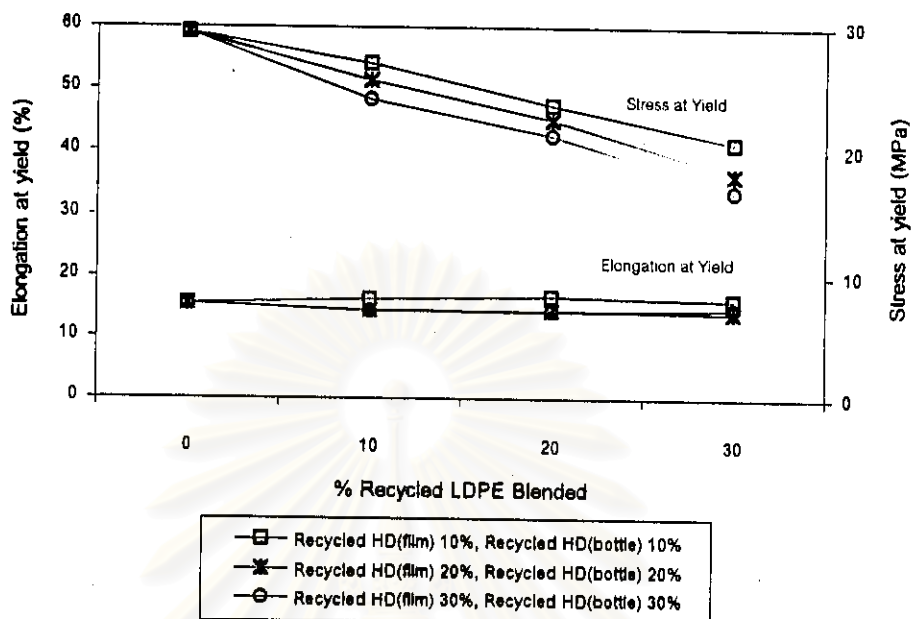
| Material (%) |                      |                        |               | $\sigma_y$ (MPa)    |                     | $\sigma_b$ (MPa)    |                     | $\epsilon_y$ (%)    |                    | $\epsilon_b$ (%)   |                    |
|--------------|----------------------|------------------------|---------------|---------------------|---------------------|---------------------|---------------------|---------------------|--------------------|--------------------|--------------------|
| Virgin HDPE  | Recycled HDPE (film) | Recycled HDPE (bottle) | Recycled LDPE | MD                  | TD                  | MD                  | TD                  | MD                  | TD                 | MD                 | TD                 |
| 100          | -                    | -                      | -             | 29.57<br>$\pm 1.76$ | 29.04<br>$\pm 0.96$ | 42.13<br>$\pm 2.34$ | 35.21<br>$\pm 1.98$ | 15.58<br>$\pm 1.16$ | 6.25<br>$\pm 0.08$ | 500<br>$\pm 11.83$ | 664<br>$\pm 35.33$ |
| 70           | 10                   | 10                     | 10            | 27.10<br>$\pm 0.59$ | 26.35<br>$\pm 0.27$ | 42.94<br>$\pm 3.24$ | 37.36<br>$\pm 2.92$ | 16.27<br>$\pm 1.41$ | 6.02<br>$\pm 0.99$ | 491<br>$\pm 37.53$ | 611<br>$\pm 32.03$ |
| 60           | 10                   | 10                     | 20            | 23.65<br>$\pm 1.18$ | 22.99<br>$\pm 1.07$ | 33.28<br>$\pm 1.48$ | 24.85<br>$\pm 0.17$ | 16.67<br>$\pm 0.36$ | 5.54<br>$\pm 0.53$ | 454<br>$\pm 22.63$ | 537<br>$\pm 40.49$ |
| 50           | 10                   | 10                     | 30            | 20.65<br>$\pm 1.02$ | 19.72<br>$\pm 0.86$ | 30.27<br>$\pm 3.30$ | 25.81<br>$\pm 1.15$ | 16.17<br>$\pm 1.32$ | 6.16<br>$\pm 0.37$ | 418<br>$\pm 17.73$ | 534<br>$\pm 39.38$ |
| 50           | 20                   | 20                     | 10            | 25.67<br>$\pm 1.22$ | 25.10<br>$\pm 0.67$ | 34.56<br>$\pm 2.72$ | 29.57<br>$\pm 2.03$ | 14.46<br>$\pm 1.53$ | 5.94<br>$\pm 0.56$ | 457<br>$\pm 17.76$ | 609<br>$\pm 20.97$ |
| 40           | 20                   | 20                     | 20            | 22.50<br>$\pm 1.57$ | 21.36<br>$\pm 1.18$ | 30.02<br>$\pm 3.90$ | 22.50<br>$\pm 2.32$ | 14.41<br>$\pm 1.18$ | 6.32<br>$\pm 1.18$ | 420<br>$\pm 19.00$ | 526<br>$\pm 15.29$ |
| 30           | 20                   | 20                     | 30            | 18.49<br>$\pm 0.43$ | 17.36<br>$\pm 0.94$ | 26.42<br>$\pm 0.33$ | 19.84<br>$\pm 0.96$ | 14.00<br>$\pm 1.42$ | 6.59<br>$\pm 0.37$ | 404<br>$\pm 34.69$ | 489<br>$\pm 30.92$ |
| 30           | 30                   | 30                     | 10            | 24.16<br>$\pm 1.16$ | 23.89<br>$\pm 1.14$ | 25.88<br>$\pm 5.26$ | 26.66<br>$\pm 2.19$ | 14.35<br>$\pm 1.27$ | 6.61<br>$\pm 0.74$ | 408<br>$\pm 28.12$ | 535<br>$\pm 26.69$ |
| 20           | 30                   | 30                     | 20            | 21.20<br>$\pm 0.89$ | 20.51<br>$\pm 1.72$ | 23.46<br>$\pm 1.80$ | 16.46<br>$\pm 2.43$ | 14.28<br>$\pm 4.57$ | 5.52<br>$\pm 0.47$ | 384<br>$\pm 7.56$  | 462<br>$\pm 14.74$ |
| 10           | 30                   | 30                     | 30            | 16.85<br>$\pm 0.76$ | 15.39<br>$\pm 1.29$ | 20.98<br>$\pm 0.60$ | 14.92<br>$\pm 1.15$ | 14.70<br>$\pm 0.98$ | 6.44<br>$\pm 0.12$ | 366<br>$\pm 26.89$ | 399<br>$\pm 50.29$ |



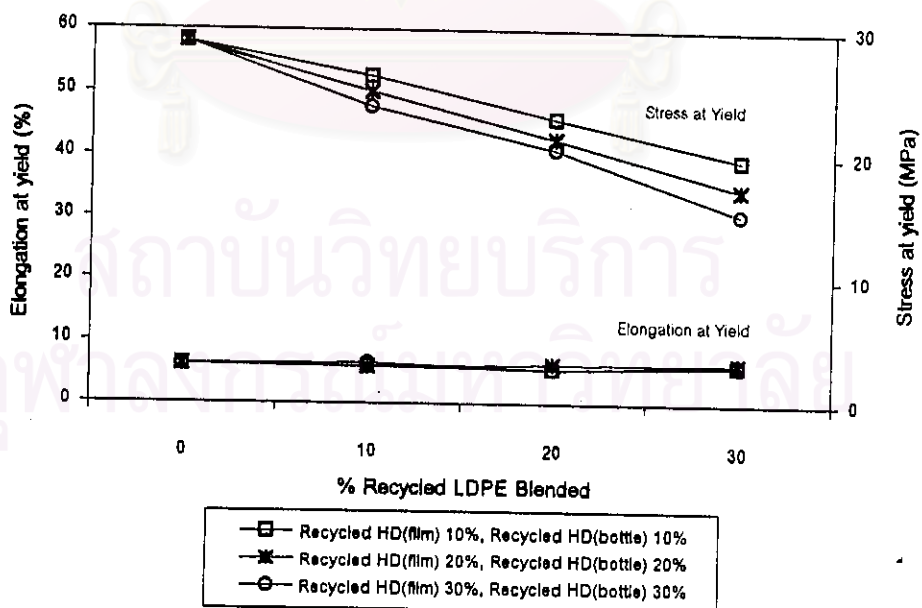
**Figure 4.2** Effect of percent recycled LDPE content on film properties (machine direction) of a 20- $\mu\text{m}$  blown film made from the virgin HDPE/recycled PE blend.



**Figure 4.3** Effect of percent recycled LDPE content on film properties (transverse direction) of a 20- $\mu\text{m}$  blown film made from the virgin HDPE/recycled PE blend.



**Figure 4.4** Effect of percent recycled LDPE content on film properties (machine direction) of a 20- $\mu$ m blown film made from the virgin HDPE/recycled PE blend.



**Figure 4.5** Effect of percent recycled LDPE content on film properties (transverse direction) of a 20- $\mu$ m blown film made from the virgin HDPE/recycled PE blend.



Figures 4.2-4.5 show that the composition of blended polyethylene resins affects the film properties. The effect of recycled polyethylene resins/virgin HDPE composition on tensile properties of the blends was investigated. We used Thai Industrial Standard (TIS)[46] of PE resin for comparison with the blended films. We choose PE type 3 (HDPE;  $MFI_{190/2.16} < 0.4$  g/10 min) for comparison with our blended films. The results indicate that the stress at break, elongation at break, and stress at yield were considerably affected while the elongation at yield did not change significantly. It was found that the recycled plastic having 10 percent of recycled LDPE gave the highest stress at yield, from 22.5 to 36.3 percent, and elongations at yield and at break, from 57.2 to 83.7 percent. On the other hand, the films containing 30 percent of recycled LDPE provided the lowest stresses at yield (between 8.6-17.8 percent) and at break, and elongation at break (between 27.5-58.7 percent). As expected, those of 20 percent of recycled LDPE gave the middle values (stress at yield from 6.4 to 18.9 percent, and elongation at break from 41.0 to 65.2 percent) of tensile property. This might be explained in terms of thermal degradation of the recycled LDPE. Lower molecular weights of the recycled LDPE resin which occurred during processing at elevated temperature may involve either oxidation or thermal degradation. On exposure to sunlight or high energy radiation, either the recycled LDPE itself or impurities in the resins absorb the radiation which induces degradation reactions resulting in a loss of tensile property. In the case of exposing to the high energy radiation, the polymer chains can split directly. In a mechanical testing, chain splitting could occur when a load or a force is given to the recycled LDPE films.

Therefore, the stress and elongation at break, and the stress at yield decreased while the elongation at yield did not change very much. The more the recycled LDPE resin, the greater the lower molecular weight and the broader the MWD as a bimodal pattern. Influence of the amorphous region of the recycled

LDPE resin overwhelms the strength of the crystalline region, which could possibly deform the polymer matrix.

#### 4.1.3 Effect on Viscoelastic Properties.

The Dynamic Mechanical Analysis (DMA) is the measurement of viscoelastic properties of polymer which relates to its chemical structure. Most polymers show behavior intermediately between an elastic solid and a viscous liquid; and such material property is classified as viscoelastic. When a polymer was degraded via a thermal degradation, and/or an oxidation, and/or high energy radiation, the polymer chains are broken or chain scission occur leading to a decrease in molecular weight. Likewise, the broken, small chains can recombine to give a longer molecular chain leading to the increase in molecular weight.

Virgin HDPE, recycled HDPE (film), recycled HDPE (bottle), recycled LDPE, and virgin HDPE blended with recycled polyethylene resins were subjected to DMA measurement to observe the change of their viscoelastic properties. The results are shown in Table 4.5 and Figures 4.6-4.10.

Young's storage moduli ( $E'$ ) of the virgin HDPE, recycled HDPE (film), recycled HDPE (bottle), and recycled LDPE, the  $E'$  of virgin HDPE are not much different from those of the recycled HDPE (bottle). The highest and lowest  $E'$  were found in the virgin HDPE ( $8 \times 10^9$  Pa) and recycled LDPE ( $4.2 \times 10^9$  Pa), respectively. It can be explained that the energy stored elastically of virgin HDPE may be caused by the long chain molecular structure of the high molecular weight HDPE, and the recycled HDPE (bottle) has also the similarly high storage elasticity which may come from the chain scission reaction leading to a broad molecular weight distribution (MWD). The lowest storage modulus of the recycled LDPE could possibly result from the branch structure of polymer chains.

For Young's loss modulus ( $E''$ ), it is a measure of energy lost as heat. Figure 4.7 shows that the highest Young's loss modulus was found in the virgin HDPE ( $6.8 \times 10^8$  Pa) and the lowest one in the recycled LDPE ( $2.2 \times 10^8$  Pa). The lower molecular weight of the polymer, the less the heat loss. We anticipated that the viscous region or the amorphous region of the polymer could dissipate the heat. Therefore, more energy lost as heat could occur and the less energy was left for measurement. The following is the sequential order of  $E''$  of the present research polymers:

$E''$  of virgin HDPE > recycled HDPE (film) ~ recycled HDPE (bottle) > recycled LDPE

Tan  $\delta$  is called the loss tangent;  $\delta$  being the angles between the in-phase and out-of-phase component in the cyclic motion. Tan  $\delta$  is the ratio of the loss modulus ( $E''$ ) to the storage modulus ( $E'$ ). Tan  $\delta$  also goes through a series of maxima. The maxima in  $E''$  and tan  $\delta$  are used as  $T_g$ . From Table 4.5 and Figures 4.8-4.10,  $T_g$ s of the virgin HDPE, recycled HDPE (film), recycled HDPE (bottle), and the blended resins were found to be approximately constant range of -118 to -119 °C, except for the recycled LDPE being shifted to -126 °C. The recycled LDPE needs more energy in a cyclic motion due to the high branching molecular structure. All the blends exhibit a single  $T_g$  which indicates that they are compatible in all proportions. We also observe that this single  $T_g$  value is in the intermediate of both polymers which possess different crystallinity.

Addition of a second component may take the form of copolymerization or polymer blending. Based on the thermodynamic theory of the glass transition, Couchman[31] derived relations to predict the  $T_g$  composition dependence of binary mixtures of miscible high polymers and other systems. The treatment that follows is easily generalized to this case for random copolymers. Theoretically, we

also can calculate  $T_g$  values of the two polymers having two pure components using the famous Fox equation:

$$\frac{1}{T_g} = \frac{M_1}{T_{g1}} + \frac{M_2}{T_{g2}} \quad (4.1)$$

or

$$\frac{1}{T_g} = \frac{W_1}{T_{g1}} + \frac{W_2}{T_{g2}} \quad (4.2)$$

Where  $M_1$  and  $M_2$  are their respective mole fractions (moles of mers for the polymers),  $W_1$  and  $W_2$  are their respective mass (weight) fractions,  $T_{g1}$  and  $T_{g2}$  their respective pure-component glass transition temperatures. For the glass transition temperature of polyethylene appeared near  $-120^\circ\text{C}$  is thought to involve the Schatzki mechanism [31]. The calculated  $T_g$ s are shown in Table 4.5.

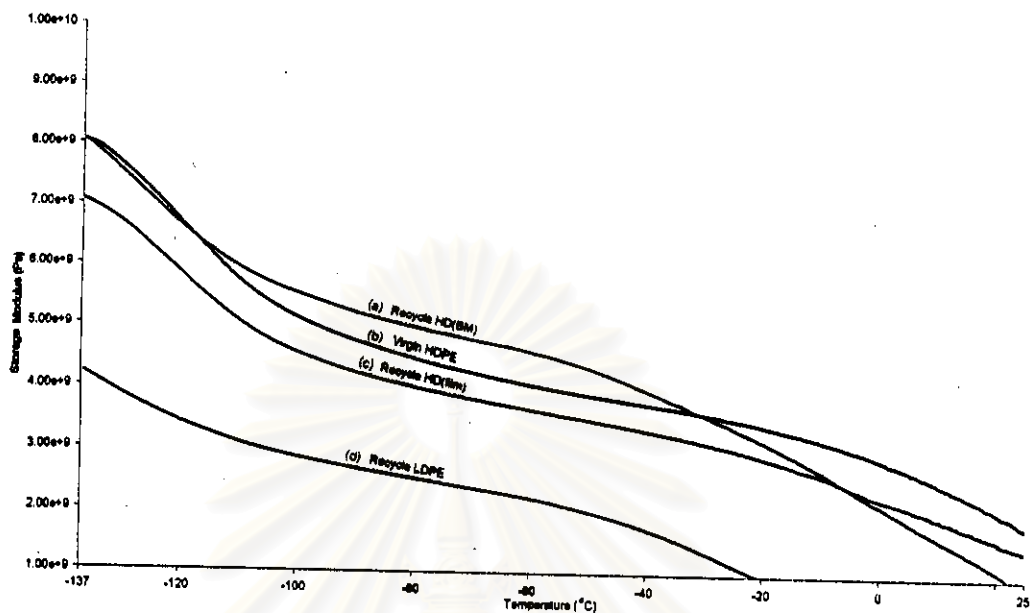
$\tan \delta$  of recycled HDPE (bottle) > virgin HDPE > recycled HDPE (film) > recycled LDPE

Basically, there is a few branching units in the virgin HDPE. The low branching molecular structure at low molecular weight of recycled HDPE (bottle grade) can be easily move. For the recycled HDPE (film), the high molecular weight ( $MFI = 0.60 \pm 0.01$ ) and high branching could be quite rigid. The recycled LDPE has a lot of branching molecular structure, therefore, the energy for chain movement is high, and the loss energy is the lowest of all.

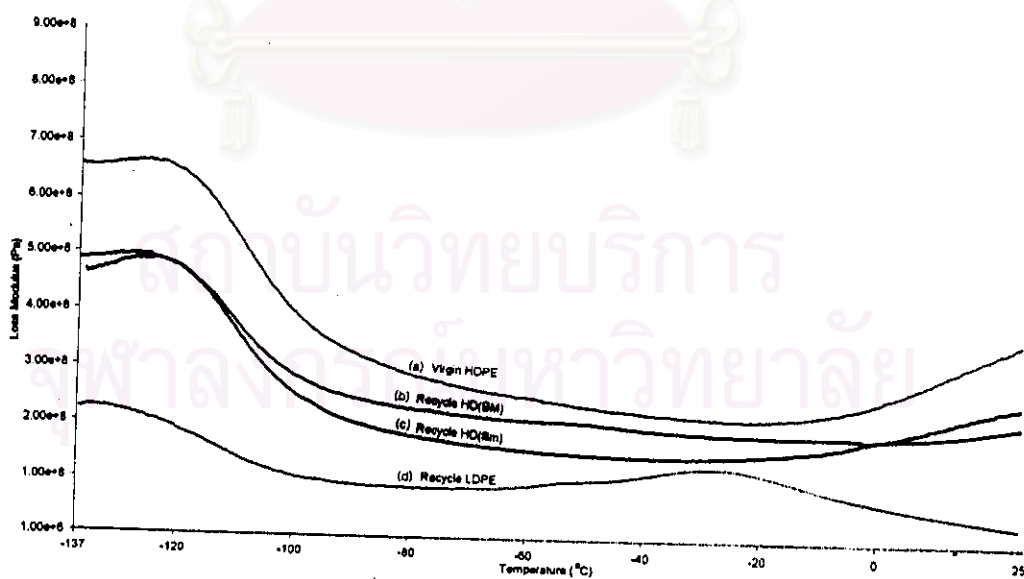
For the blends as shown in Figures 4.9-4.11, we can see the different effects on MFI and viscoelasticity. As mentioned previously, the recycled LDPE has a substantial amount of branching units, which cause the rigid molecular chains and a sluggish backbone movement. It needs an excess amount of energy to make it flow. Thus, the blends of the recycled LDPE need higher energy for chain movement, flow and processing.

**Table 4.5** The Young's Storage Moduli ( $E'$ ), Loss Moduli ( $E''$ ),  $T_g$ , and  $\tan \delta$  of Virgin HDPE Blended with Recycled PE Resins.

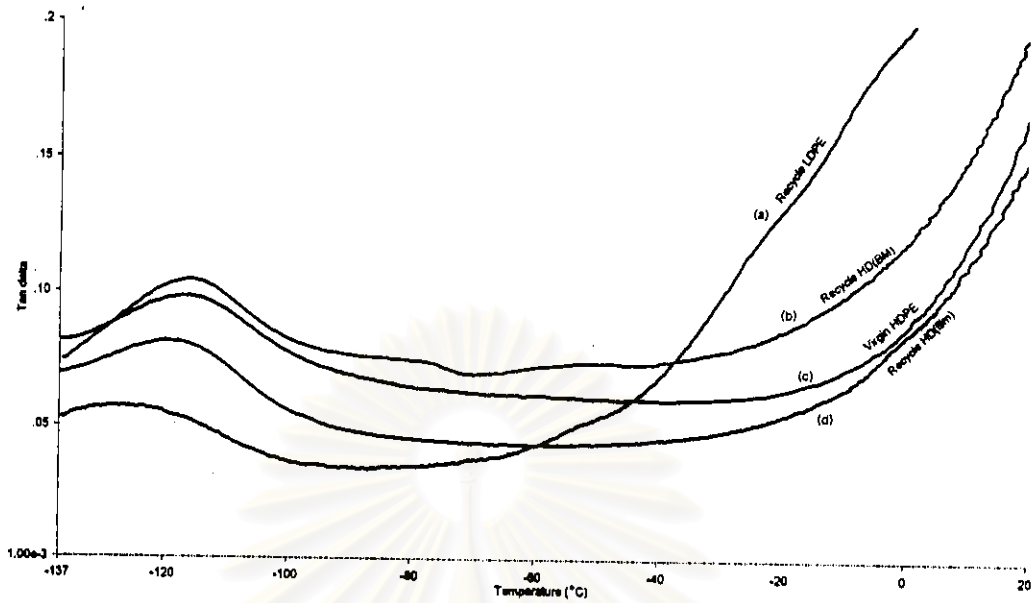
| Material (%)   |                            |                              |                  | MFL <sub>2.16</sub><br>(g/10min) | $E' \times 10^{-9}$<br>at 137 °C<br>(Pa) | $E'' \times 10^{-8}$<br>at 137 °C<br>(Pa) | $T_g$<br>(measured)<br>(°C) | $T_g$<br>(calculated)<br>(°C) | Tan $\delta$ |
|----------------|----------------------------|------------------------------|------------------|----------------------------------|--|---|-----------------------------|-------------------------------|--------------|
| Virgin<br>HDPE | Recycled<br>HDPE<br>(film) | Recycled<br>HDPE<br>(bottle) | Recycled<br>LDPE |                                  |  |   |                             |                               |              |
| 100            | -                          | -                            | -                | 0.06±0.01                        | 8.0                                      | 6.8                                       | -119.0                      | -                             | 0.098        |
| -              | 100                        | -                            | -                | 0.06±0.01                        | 7.1                                      | 4.7                                       | -118.5                      | -                             | 0.081        |
| -              | -                          | 100                          | -                | 2.50±0.03                        | 8.0                                      | 4.9                                       | -116.7                      | -                             | 0.104        |
| -              | -                          | -                            | 100              | 4.30±0.07                        | 4.2                                      | 2.2                                       | -126.0                      | -                             | 0.057        |
| 70             | 10                         | 10                           | 10               | 0.07±0.01                        | 6.7                                      | 4.6                                       | -117.7                      | -120                          | 0.092        |
| 60             | 10                         | 10                           | 20               | 0.15±0.01                        | 6.2                                      | 3.3                                       | -119.7                      | -120                          | 0.074        |
| 50             | 10                         | 10                           | 30               | 0.17±0.02                        | 4.7                                      | 2.7                                       | -119.4                      | -120                          | 0.082        |
| 30             | 30                         | 30                           | 10               | 0.18±0.01                        | 6.3                                      | 3.9                                       | -117.9                      | -120                          | 0.063        |
| 20             | 30                         | 30                           | 20               | 0.30±0.01                        | 6.0                                      | 2.8                                       | -118.4                      | -120                          | 0.067        |
| 10             | 30                         | 30                           | 30               | 0.47±0.01                        | 5.3                                      | 2.7                                       | -119.4                      | -120                          | 0.088        |



**Figure 4.6** Young's storage moduli of the virgin HDPE and the recycled PE resins: (a) recycled HDPE (bottle); (b) virgin HDPE; (c) recycled HDPE (film); (d) recycled LDPE.

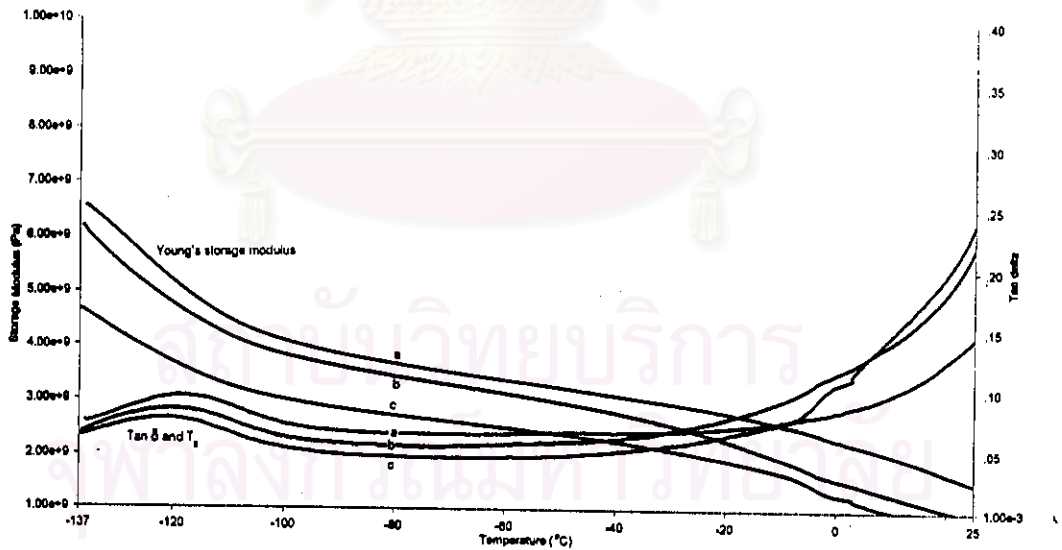


**Figure 4.7** Young's loss moduli of the virgin HDPE and the recycled PE resins: (a) virgin HDPE; (b) recycled HDPE (bottle); (c) recycled HDPE (film); (d) recycled LDPE.



**Figure 4.8**  $T_g$  and  $\tan \delta$  of the virgin HDPE and the recycled PE resins:

(a) recycled LDPE; (b) recycled HDPE (bottle); (c) virgin HDPE; (d) recycled HDPE (film).

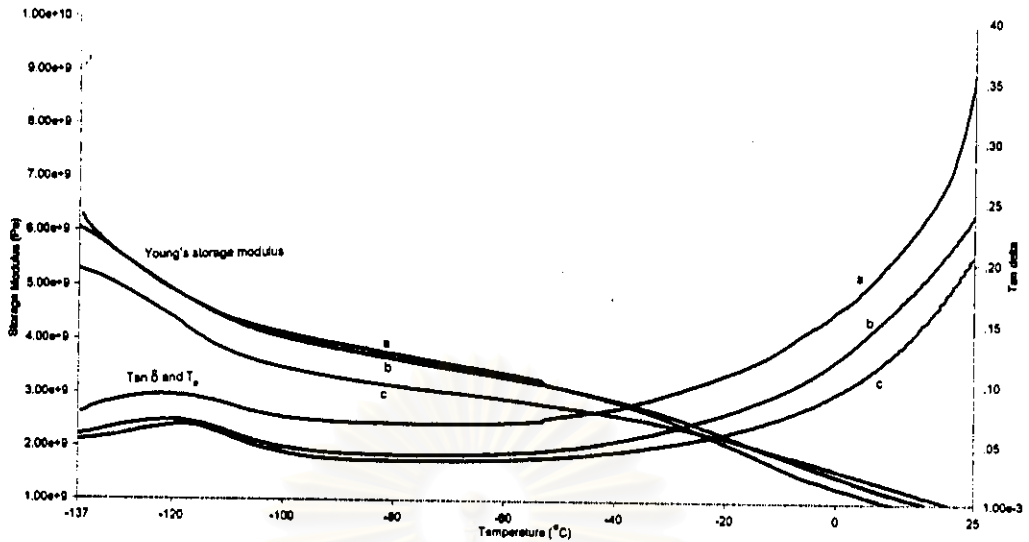


**Figure 4.9** Young's storage modulus,  $\tan \delta$ , and  $T_g$  of the blended PE resins:

(a) virgin HDPE 70%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 10%;

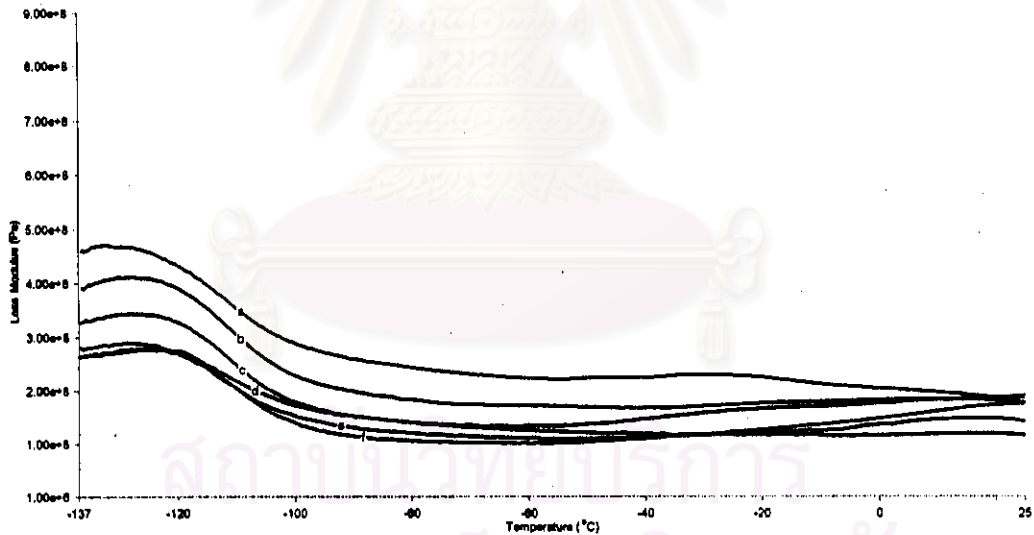
(b) virgin HDPE 60%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 20%;

(c) virgin HDPE 50%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 30%.



**Figure 4.10** Young's storage moduli,  $\tan \delta$ , and  $T_g$  of the blended PE resins:

- (a) virgin HDPE 30%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 10%;
- (b) virgin HDPE 20%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 20%;
- (c) virgin HDPE 10%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 30%.



**Figure 4.11** Young's loss moduli of the blended PE resins:

- (a) virgin HDPE 70%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 10%;
- (b) virgin HDPE 30%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 10%;
- (c) virgin HDPE 60%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 20%;
- (d) virgin HDPE 30%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 20%;
- (e) virgin HDPE 50%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 30%;
- (f) virgin HDPE 10%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 30%.



## 4.2 Effect of Virgin HDPE and Recycled Polyethylene Compositions on Rheological Properties.

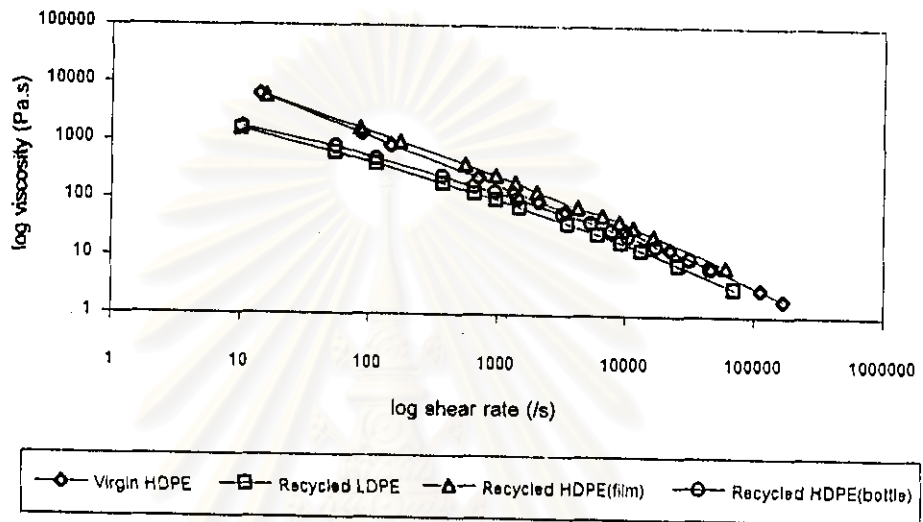
For a molten polymer, each rheological property provides a different basis for comparison and characterization. Comparisons of the properties of virgin HDPE, recycled polyethylene resins, and a series of virgin HDPE/recycled polyethylene resin blends were studied in terms of melt viscosity.

The flow curves of virgin HDPE, recycled polyethylene resins, and virgin HDPE/recycled polyethylene resin blends are shown in Figures 4.12-4.15. As expected, log viscosity ( $\eta$ ) decreases with increasing log shear rate ( $\dot{\gamma}$ ) at a constant temperature (190 °C) as shown in Figure 4.12. In order to improve the properties of the recycled polyethylene resins, they were blended among themselves. The results in Figures 4.13-4.15 show that a high proportion of blended recycled LDPE decreased the viscosity because the recycled LDPE has high contents of low molecular weight fraction, which give less resistance to flow. From flow curves in Figures 4.13-4.15, there is not much difference in viscosity at a specific shear rate. The Newtonian melt viscosity,  $\eta_0$ , of a Newtonian material depends upon the weight average molecular weight,  $\bar{M}_w$ , [1] as follows:

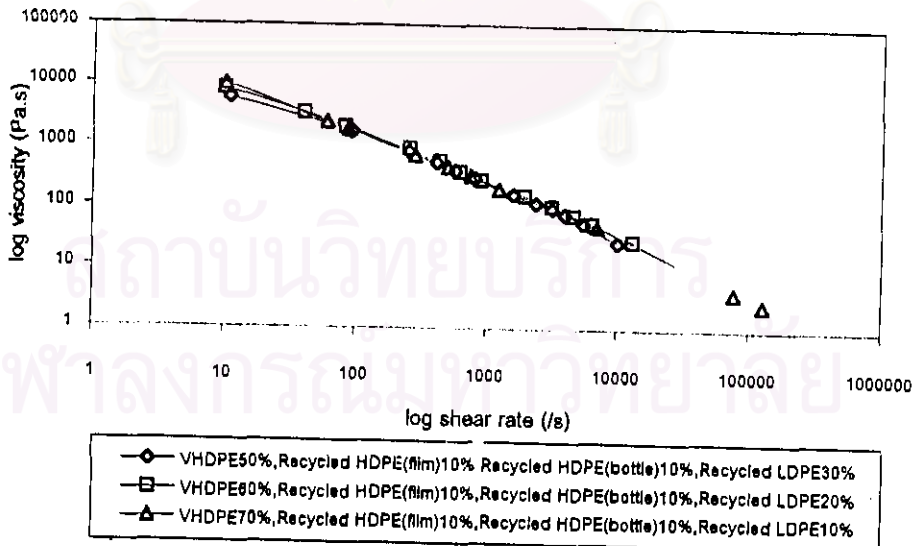
$$\eta_0 = K \bar{M}_w^{3.4-3.5} \quad (4.3)$$

where K is the consistency of melt viscosity. The non-Newtonian melt viscosity (as our case) and the shape of the flow curve depend on the weight-average molecular weight, as well as on the molecular weight distribution (MWD). Most film industries use recycled LDPE resin for blending with either the virgin HDPE or recycled HDPE resin because the virgin/recycled LDPE resin gives good processability and material saving. When we use a large amount of HDPE or

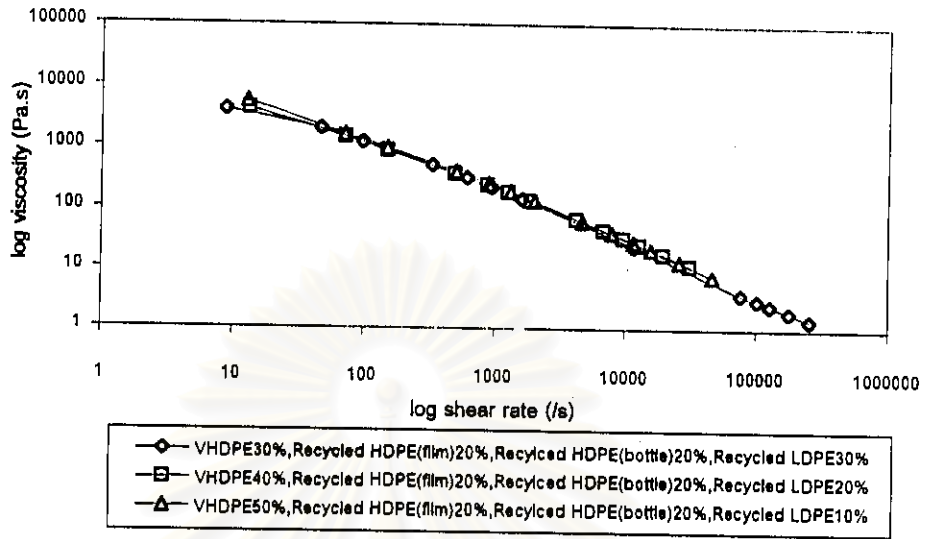
HDPE alone, increase in melt viscosity usually introduces processing problems since HDPE has high molecular weights and a narrow molecular weight distribution. The blends of HDPE and LDPE or recycled LDPE help improve the film processability at a high shear rate and prolong the lifetime of the extruder.



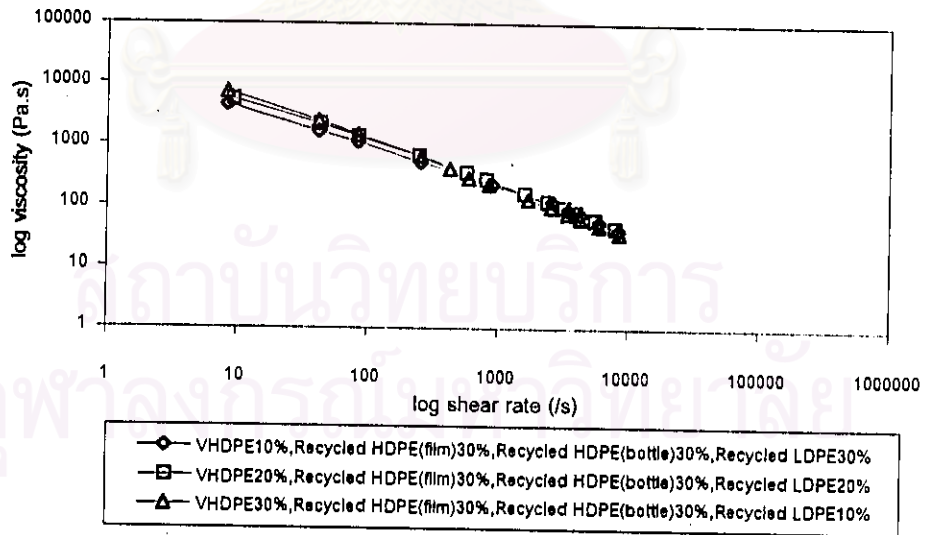
**Figure 4.12** Flow curves of the virgin HDPE, and the recycled PE resins.



**Figure 4.13** Flow curves of the PE blended resins: (a constant amount of the recycled HDPE (film) and recycled HDPE (bottle) at 10 percent with various amounts of recycled LDPE).



**Figure 4.14** Flow curves of the PE blended resins: (a constant amount of the recycled HDPE (film) and recycled HDPE (bottle) at 20 percent with various amounts of recycled LDPE).



**Figure 4.15** Flow curves of the PE blended resins: (a constant amount of the recycled HDPE (film) and recycled HDPE (bottle) at 30 percent with various amounts of recycled LDPE).

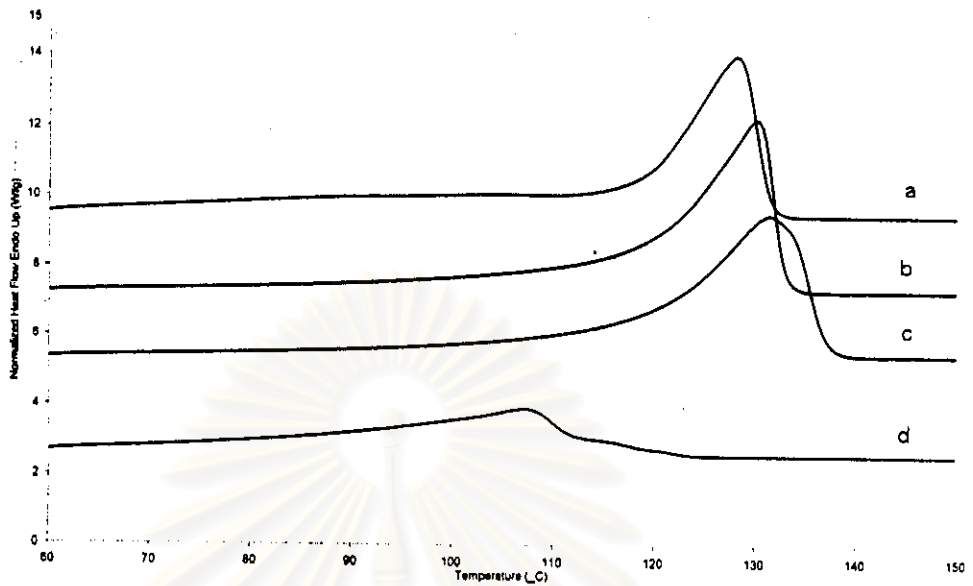
### 4.3 Effect of Virgin HDPE and Recycled Polyethylene Compositions on Thermal Characteristics.

#### 4.3.1 Melting Behavior.

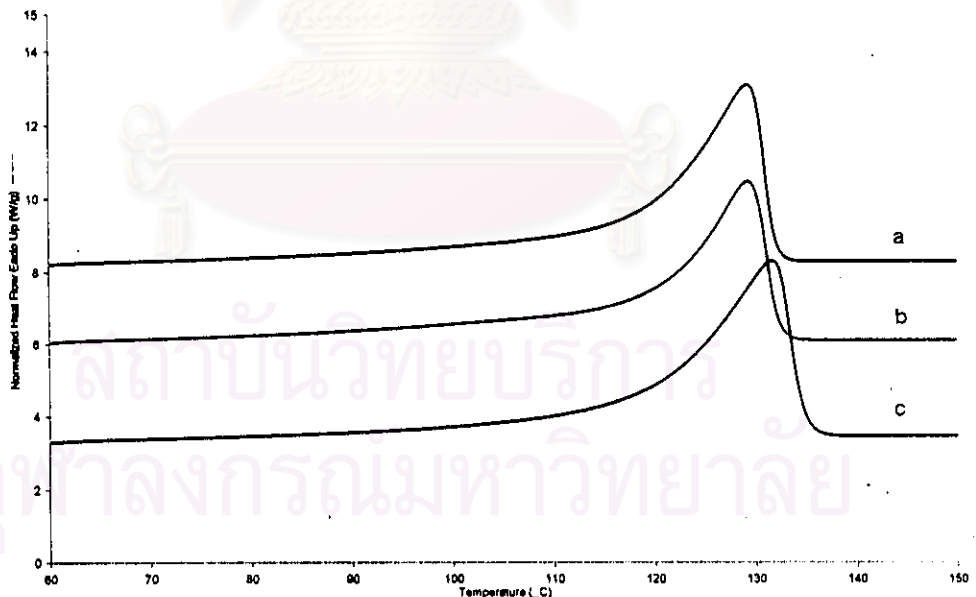
The melting behavior of the blended polymers was measured by DSC. The melting point and heat of fusion for all resins are summarized in Table 4.6 and shown in Figures 4.16-4.19.

**Table 4.6** Melting Characteristics of PE Resins and the Blended Resins.

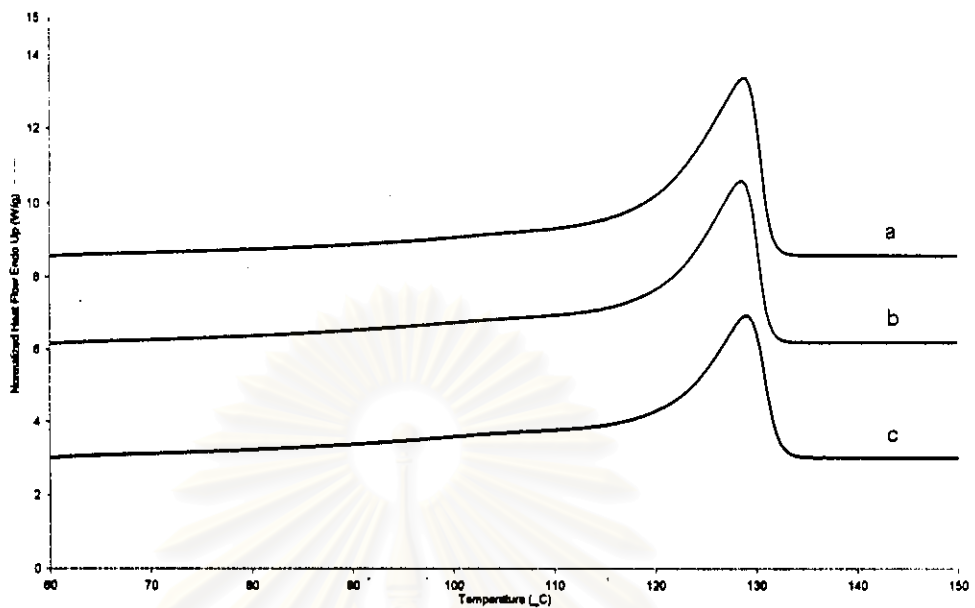
| Material (%)   |                         |                           |               | T <sub>m</sub><br>(°C) | Heat of Fusion<br>(J/g) |
|----------------|-------------------------|---------------------------|---------------|------------------------|-------------------------|
| Virgin<br>HDPE | Recycled HDPE<br>(film) | Recycled HDPE<br>(bottle) | Recycled LDPE |                        |                         |
| 100            | -                       | -                         | -             | 131.4                  | 184.7                   |
| -              | 100                     | -                         | -             | 130.1                  | 178.4                   |
| -              | -                       | 100                       | -             | 128.1                  | 157.7                   |
| -              | -                       | -                         | 100           | 107.1                  | 91.1                    |
| 70             | 10                      | 10                        | 10            | 129.1                  | 183.5                   |
| 60             | 10                      | 10                        | 20            | 129.4                  | 170.7                   |
| 50             | 10                      | 10                        | 30            | 131.7                  | 170.5                   |
| 50             | 20                      | 20                        | 10            | 128.7                  | 174.0                   |
| 40             | 20                      | 20                        | 20            | 128.4                  | 170.5                   |
| 30             | 20                      | 20                        | 30            | 129.1                  | 160.4                   |
| 30             | 30                      | 30                        | 10            | 129.4                  | 163.3                   |
| 20             | 30                      | 30                        | 20            | 129.1                  | 158.4                   |
| 10             | 30                      | 30                        | 30            | 127.4                  | 146.1                   |



**Figure 4.16** DSC thermograms showing the melting points of virgin HDPE and recycled polyethylene resins: (a) recycled HDPE (bottle); (b) recycled HDPE (film); (c) virgin HDPE; (d) recycled LDPE.

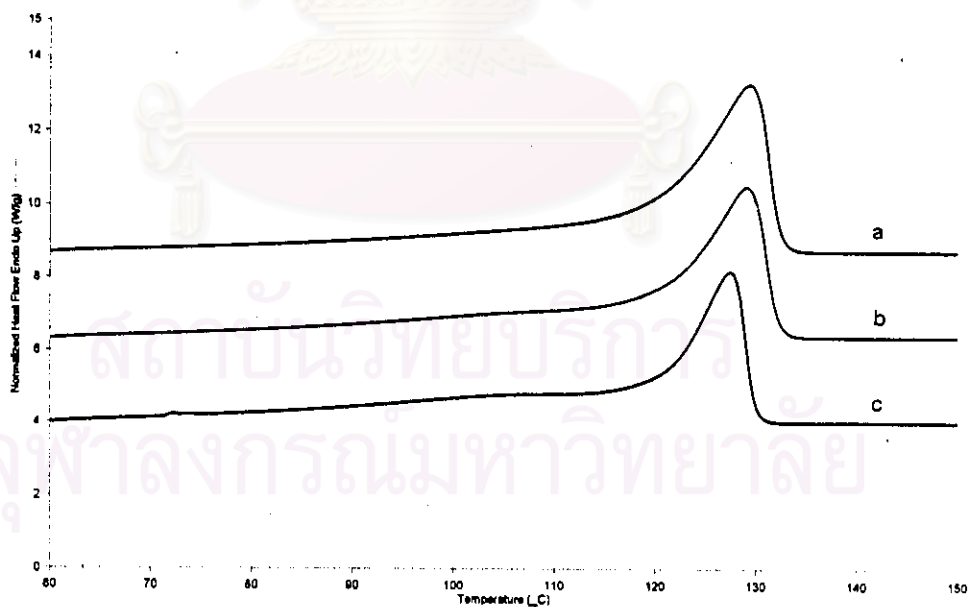


**Figure 4.17** DSC thermograms showing the melting points of the blended resins: (a) virgin HDPE 70%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 10%; (b) virgin HDPE 60%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 20%; (c) virgin HDPE 50%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 30%.



**Figure 4.18** DSC thermograms showing the melting points of the blended resins:

- (a) virgin HDPE 50%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 10%;
- (b) virgin HDPE 40%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 20%;
- (c) virgin HDPE 30%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 30%.



**Figure 4.19** DSC thermograms showing the melting points of the blended resins:

- (a) virgin HDPE 30%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 10%;
- (b) virgin HDPE 20%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 20%;
- (c) virgin HDPE 10%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 30%.

For the melting points ( $T_m$ ) of virgin HDPE, recycled HDPE (film), and recycled HDPE (bottle), recycled LDPE, and the blends, a single melting peak was observed. It indicates that all phases in each blend were compatible at all compositions. The melting points of HDPE (128-131 °C) differs from that of LDPE (107 °C), but the melting points of blended PE resins are not significantly different. The heat of fusion shows a better correlation between composition and crystallinity, which shall be discussed in Section 4.5.

#### **4.4 Effect of Virgin HDPE and Recycled Polyethylene Compositions on Morphological Characteristics.**

##### **4.4.1 Scanning Electron Microscopy (SEM).**

Scanning electron microscope (SEM) was employed to investigate the morphology of the blended recycled polyethylene films. As mentioned in Chapter 3, they were etched by  $KMnO_4$  in a mixture of concentrated sulphuric acid and phosphoric acid. The morphology of the blends was shown in Figures 4.20-4.32.

The lamellar structure of films is well shown in the higher magnification (15,000X) micrographs taken by zooming up around the well-developed lamellar stacks. The virgin HDPE, the recycled HDPE (film), and the recycled HDPE (bottle) have an obviously by stacked lamellar morphology, as shown in Figures 4.20-4.22. These regions are apparently more resistant to the etching solvent causing them to stand out from the film surface. The lamellar stack orientation did not change between them and was highly ordered. They demonstrated that high molecular weight components contribute to the formation of extended chain fibrils which constitute the backbone. However, the lamellar stacks of the recycled LDPE (Figure 4.23) show the network structure of low-ordercd orientation, because the

recycled LDPE has low molecular weights with the high-branching molecular structure.

The film surfaces of three different types of the recycled polyethylene blends were studied as shown in Figures 4.24-4.32. It is well-known that the morphology of the blends is greatly affected by the recycled LDPE. We can see that the film surface is not smooth with an increase of the recycled LDPE content, but the film is highly transparency compared with the virgin HDPE. This sharp appearance of the recycled LDPE affects the film surfaces in all blends. That is, the recycled LDPE with the high-branching molecular structure serves to reduce the crystallinity of film structure substantially when the recycled LDPE was blended with other recycled polyethylene resins. A single lamella appears as a bright line and the amorphous region between two adjacent lamellae is seen as a narrow dark line. A single lamella can be seen as a crystal step on the lamellar crystal where the crystal surface is below the film surface, as mentioned by Kim [26], [29], Tagawa [38], and Sherman [41]. The crystallinity region can be investigated by X-ray diffraction (XRD) for supporting the bright fibrils on film surface.

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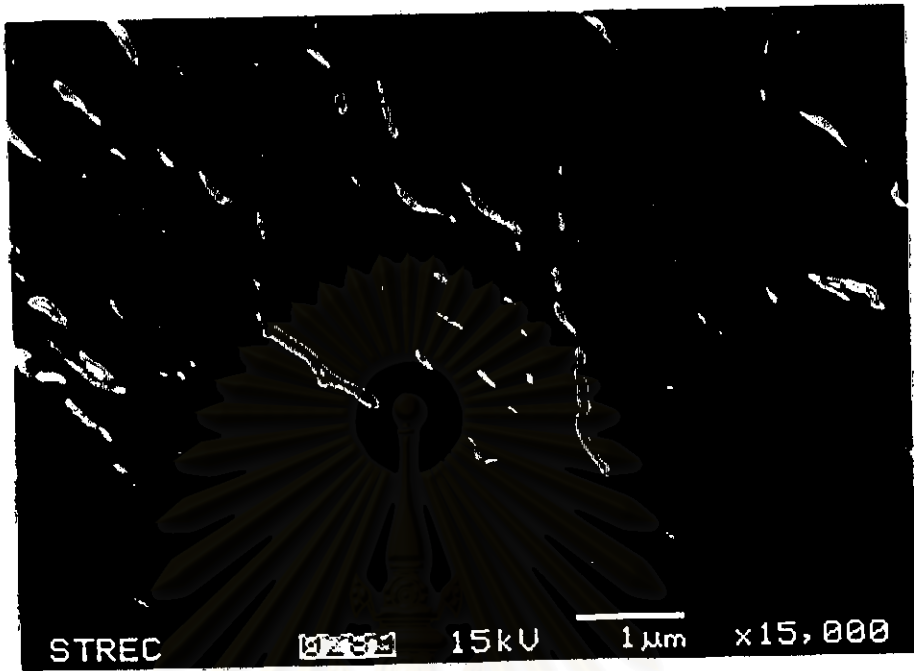


Figure 4.20 Scanning electron micrograph of the virgin HDPE film at 15,000 $\times$  magnification.

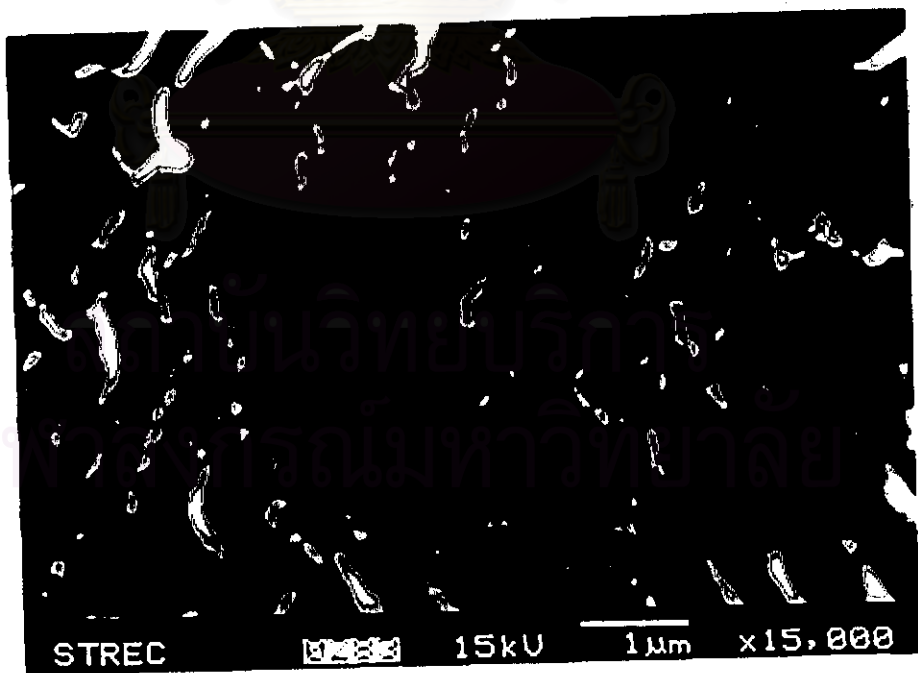


Figure 4.21 Scanning electron micrograph of the recycled HDPE (film) at 15,000 $\times$  magnification.

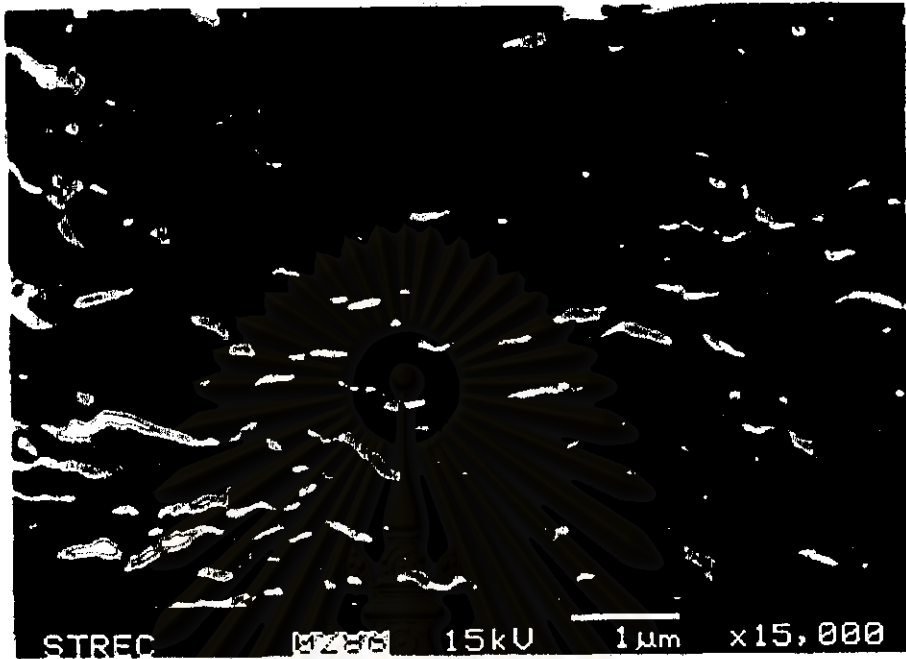


Figure 4.22 Scanning electron micrograph of the recycled HDPE (bottle) at 15,000 $\times$  magnification.



Figure 4.23 Scanning electron micrograph of the recycled LDPE film at 15,000 $\times$  magnification.

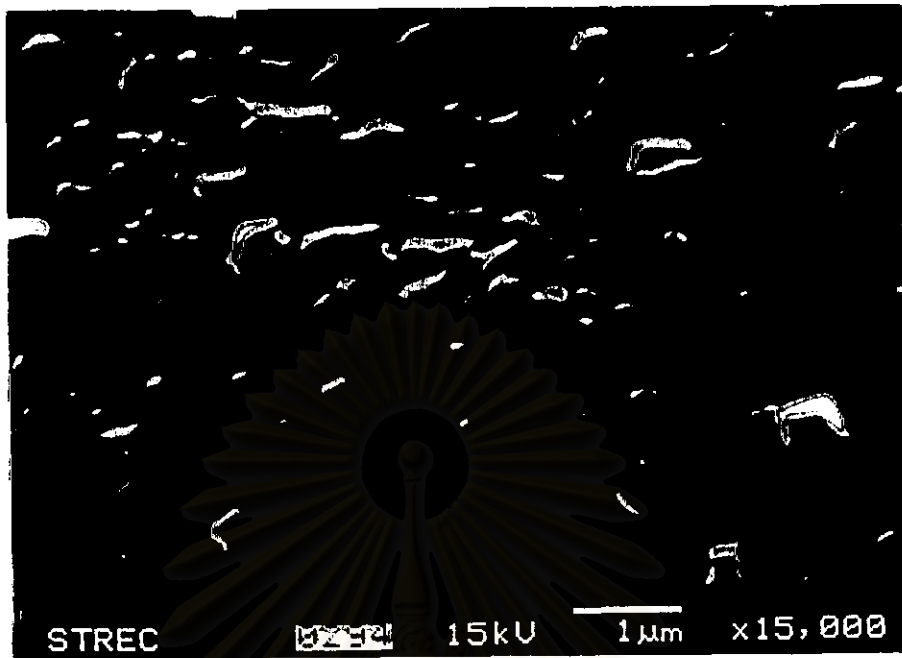


Figure 4.24 Scanning electron micrograph of the blended PE film at 15,000 $\times$  magnification (virgin HDPE 70%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 10%).

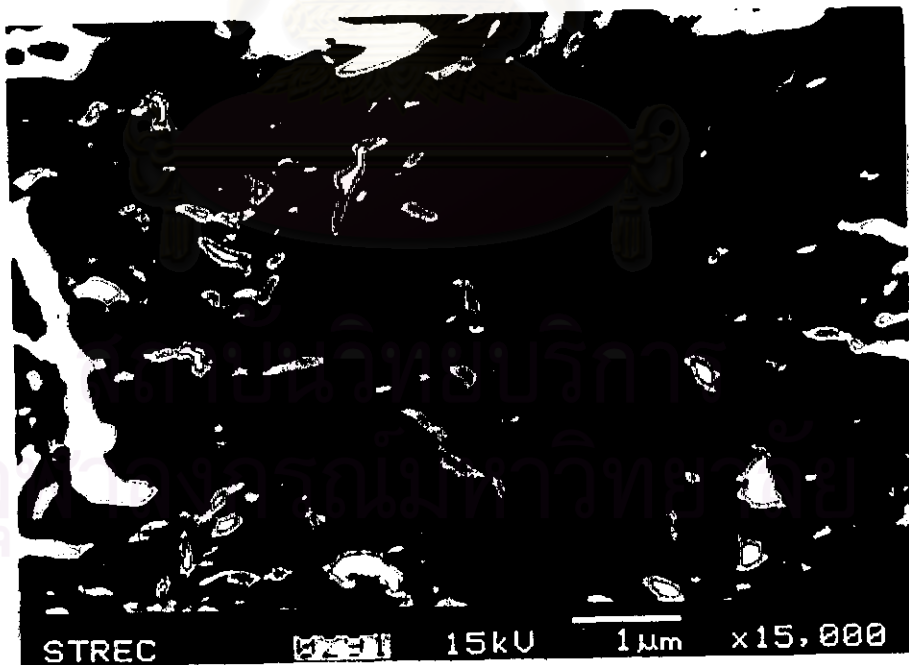


Figure 4.25 Scanning electron micrograph of the blended PE film at 15,000 $\times$  magnification (virgin HDPE 60%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 20%).

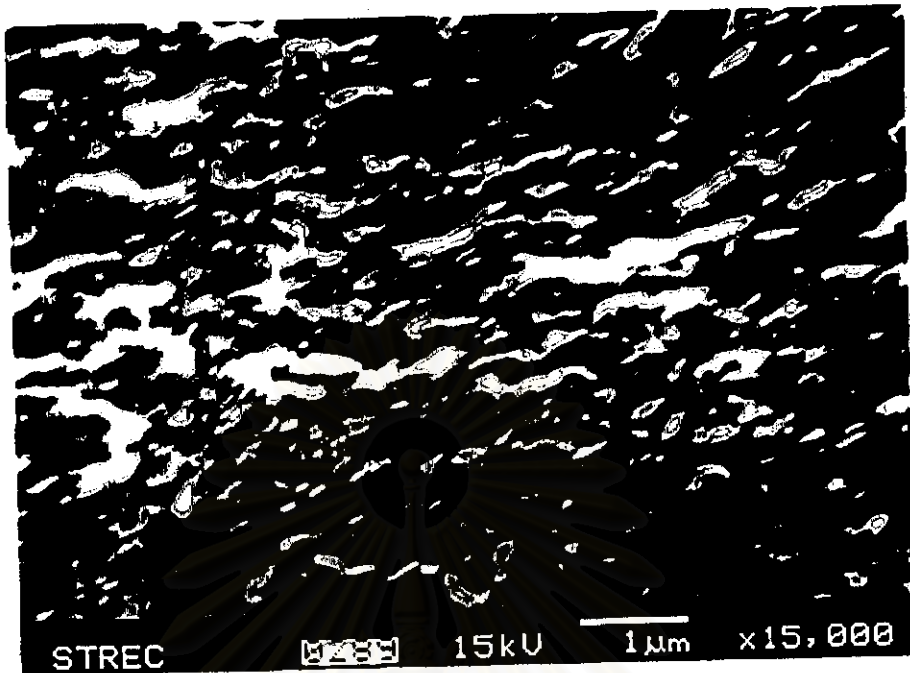


Figure 4.26 Scanning electron micrograph of the blended PE film at 15,000× magnification (virgin HDPE 50%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 30%).

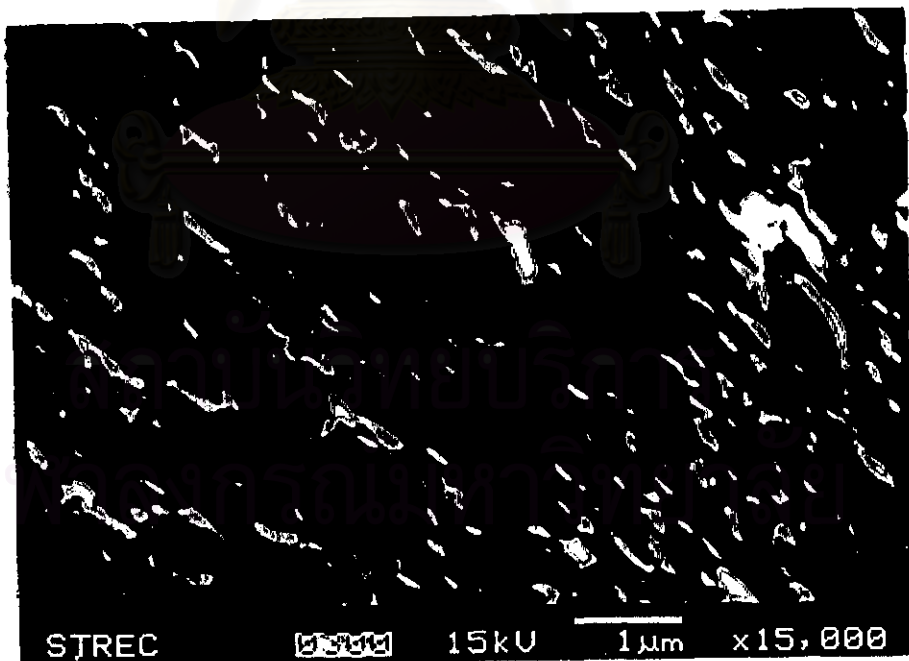
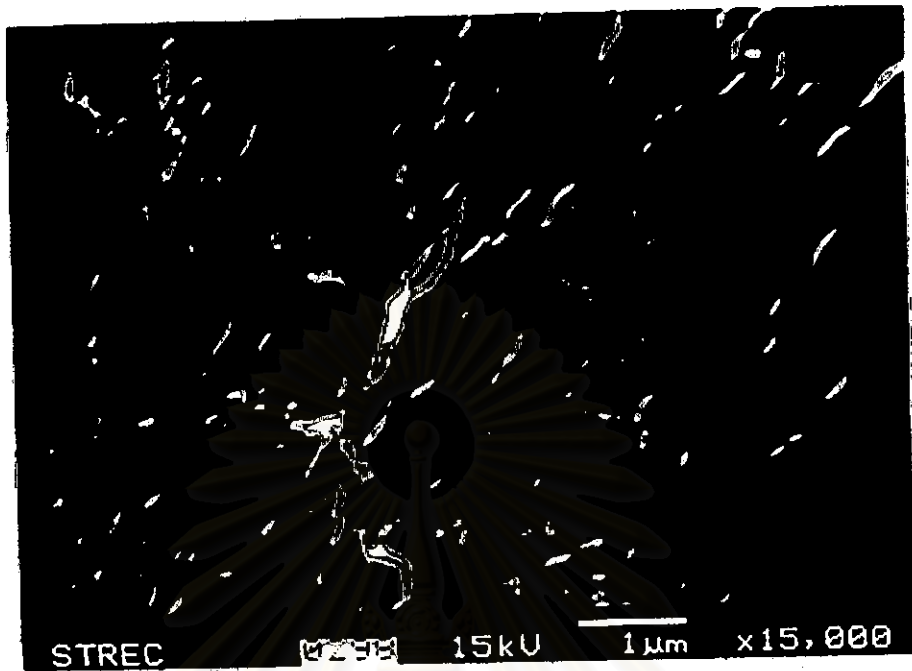
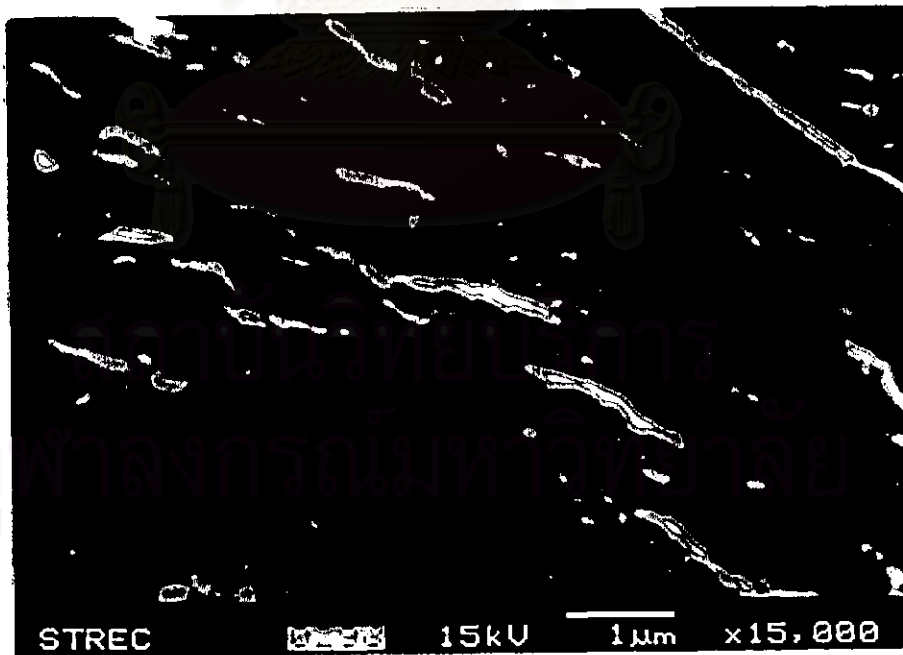


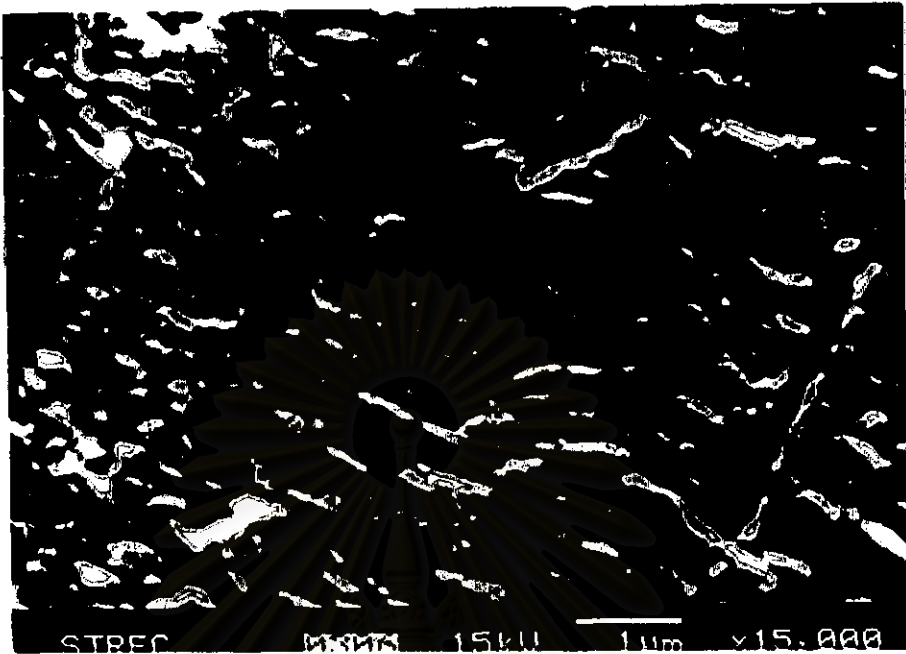
Figure 4.27 Scanning electron micrograph of the blended PE film at 15,000× magnification (virgin HDPE 50%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 10%).



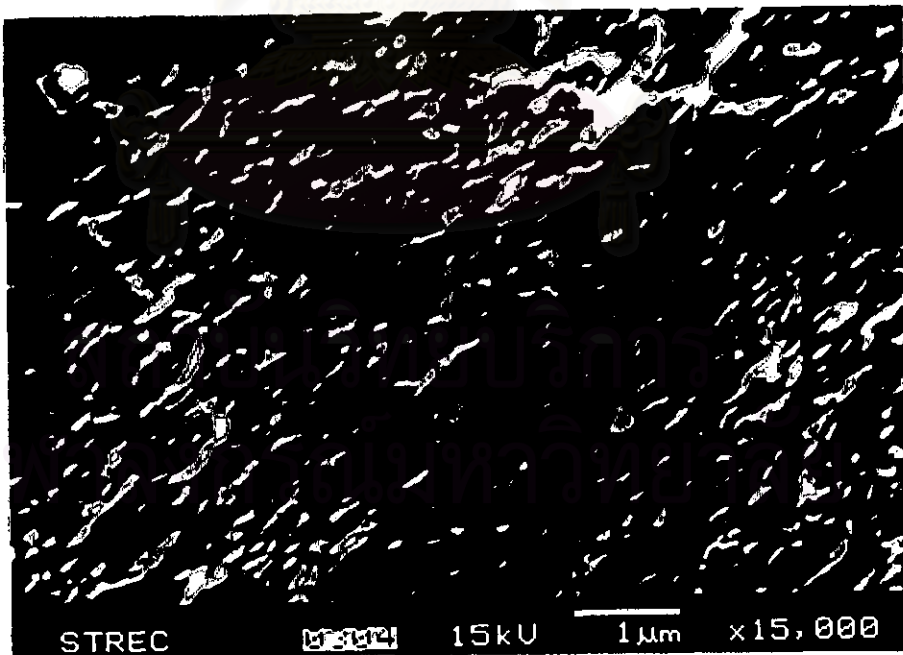
**Figure 4.28** Scanning electron micrograph of the blended PE film at 15,000 $\times$  magnification (virgin HDPE 40%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 20%).



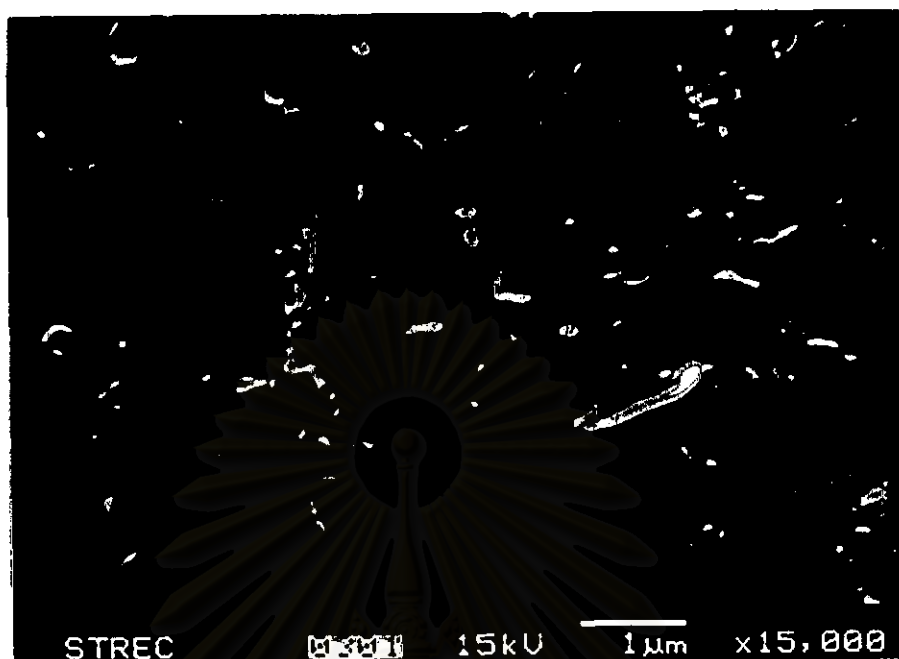
**Figure 4.29** Scanning electron micrograph of the blended PE film at 15,000 $\times$  magnification (virgin HDPE 30%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 30%).



**Figure 4.30** Scanning electron micrograph of the blended PE film at 15,000× magnification (virgin HDPE 30%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 10%).



**Figure 4.31** Scanning electron micrograph of the blended PE film at 15,000× magnification (virgin HDPE 20%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 20%).



**Figure 4.32** Scanning electron micrograph of the blended PE film at 15,000 $\times$  magnification (virgin HDPE 10%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 30%).

#### 4.5 The Degree of Crystallinity.

Two methods of analysis for percentage of crystallinity were carried out. Heat of fusion in the DSC measurement and XRD were used for the degree of crystallinity investigation.

##### 4.5.1 Differential Scanning Calorimetry (DSC).

The DSC curves of the virgin HDPE, the recycled polyethylene resins and the blends are shown in Figures 4.16-4.19. The most obvious feature is that the polymer melting peaks cover a temperature range of 127-131  $^{\circ}\text{C}$ , except that the

recycled LDPE is 107 °C. This suggests that the recycled LDPE has low molecular weights and different crystallinity compound with the recycled HDPE.

To calculate the percentage crystallinity, a value for the heat of fusion of completely (100%) crystalline polyethylene of 68.4 cal/g (287.3 J/g) was taken [42]. The polymer percent crystallinity could be calculated from eq. (4.4):

$$\% \text{ Crystallinity} = \frac{\Delta H_f \text{ of sample from DSC curve} \times 100}{\Delta H_f \text{ of theoretical 100\% crystalline sample}} \quad (4.4)$$

For DSC curves of the blends, their melting peaks resemble much more closely to that of the HDPE rather than the recycled LDPE. This suggests that the blends consist mainly of HDPE-rich crystalline regions. However, as the content of the recycled LDPE in the blends increases, the melting temperature decreases as a result of the increased chain mobility and disorder in the crystal due to the inclusion of the more flexible recycled LDPE. In other words, inclusion of more LDPE chains increases the LDPE-rich amorphous regions.

**Table 4.7** Percent Crystallinities from DSC.

| Material               | Heat of fusion <sup>a</sup><br>(J/g) | Heat of fusion <sup>b</sup><br>From DSC<br>(J/g) | Crystallinity<br>(%) |
|------------------------|--------------------------------------|--|----------------------|
| Virgin HDPE            | 287.3                                | 184.7  | 64.3                 |
| Recycled HDPE (film)   | 287.3                                | 178.4  | 62.1                 |
| Recycled HDPE (bottle) | 287.3                                | 157.7  | 54.9                 |
| Recycled LDPE          | 287.3                                | 91.1   | 31.7                 |

<sup>a</sup> is heat of fusion of a completely crystalline polyethylene, <sup>b</sup> is values read directly from DSC curves (70-140 °C).



Table 4.7 shows the percent crystallinity of HDPE and the recycled HDPE and LDPE in comparison to the heat of fusion from literature [42] and from DSC of the present work.

#### 4.5.2 X-ray Diffraction (XRD).

The X-ray diffraction curves obtained are shown in Figures 4.33-4.45 and the corresponding calculations are presented in Table 4.8.

The polymer percent crystallinity could be calculated from eq. (4.5):

$$\% \text{ Crystallinity} = \frac{\text{area under crystalline region (g)} \times 100}{\text{area under crystalline region (g)} + \text{area under amorphous region (g)}} \quad (4.5)$$

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**Table 4.8** Percent Crystallinities of Polyethylene Films from X-ray Diffraction.

| Material (%)   |                            |                              |                  | Area under<br>Crystalline<br>Region*<br>(g) | Area under<br>Amorphous<br>Region*<br>(g) | Crystallinity<br>(%) |
|----------------|----------------------------|------------------------------|------------------|---|---|----------------------|
| Virgin<br>HDPE | Recycled<br>HDPE<br>(film) | Recycled<br>HDPE<br>(bottle) | Recycled<br>LDPE |   |   |                      |
| 100            | -                          | -                            | -                | 0.038                                       | 0.011                                     | 77.6                 |
| -              | 100                        | -                            | -                | 0.033                                       | 0.011                                     | 75.0                 |
| -              | -                          | 100                          | -                | 0.033                                       | 0.013                                     | 71.7                 |
| -              | -                          | -                            | 100              | 0.017                                       | 0.016                                     | 51.5                 |
| 70             | 10                         | 10                           | 10               | 0.042                                       | 0.014                                     | 75.0                 |
| 60             | 10                         | 10                           | 20               | 0.031                                       | 0.012                                     | 72.1                 |
| 50             | 10                         | 10                           | 30               | 0.029                                       | 0.012                                     | 70.7                 |
| 50             | 20                         | 20                           | 10               | 0.029                                       | 0.010                                     | 74.4                 |
| 40             | 20                         | 20                           | 20               | 0.031                                       | 0.011                                     | 73.8                 |
| 30             | 20                         | 20                           | 30               | 0.033                                       | 0.014                                     | 70.2                 |
| 30             | 30                         | 30                           | 10               | 0.032                                       | 0.013                                     | 71.1                 |
| 20             | 30                         | 30                           | 20               | 0.032                                       | 0.014                                     | 69.5                 |
| 10             | 30                         | 30                           | 30               | 0.029                                       | 0.014                                     | 67.4                 |

\*The area is determined using a cut-and-weight method (using a filter paper with a diameter of 100 mm produced by Schleicher&Schuell, Dassel, Germany)

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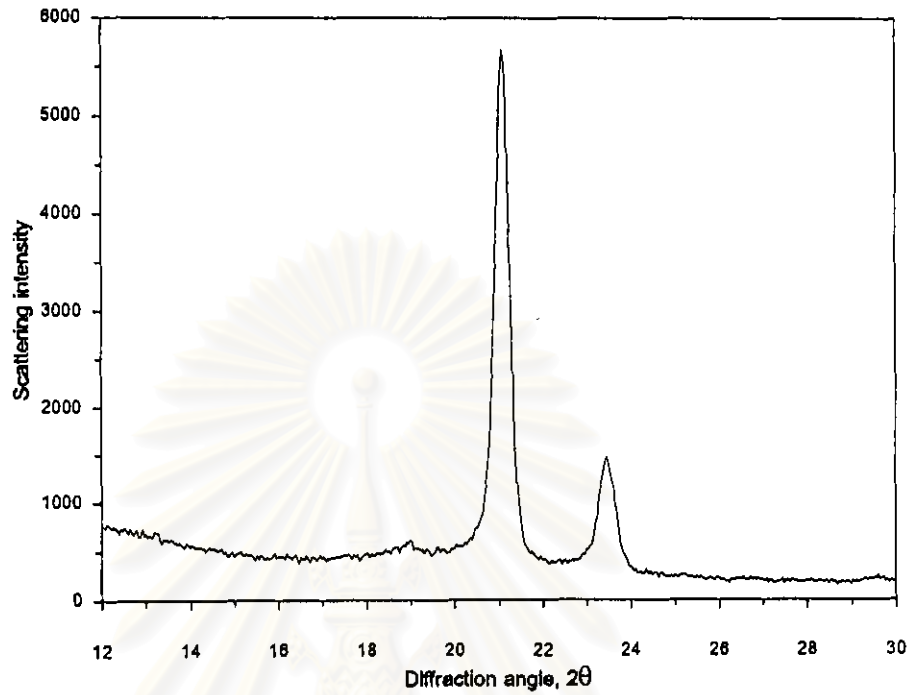


Figure 4.33 X-ray diffraction curve of the virgin HDPE.

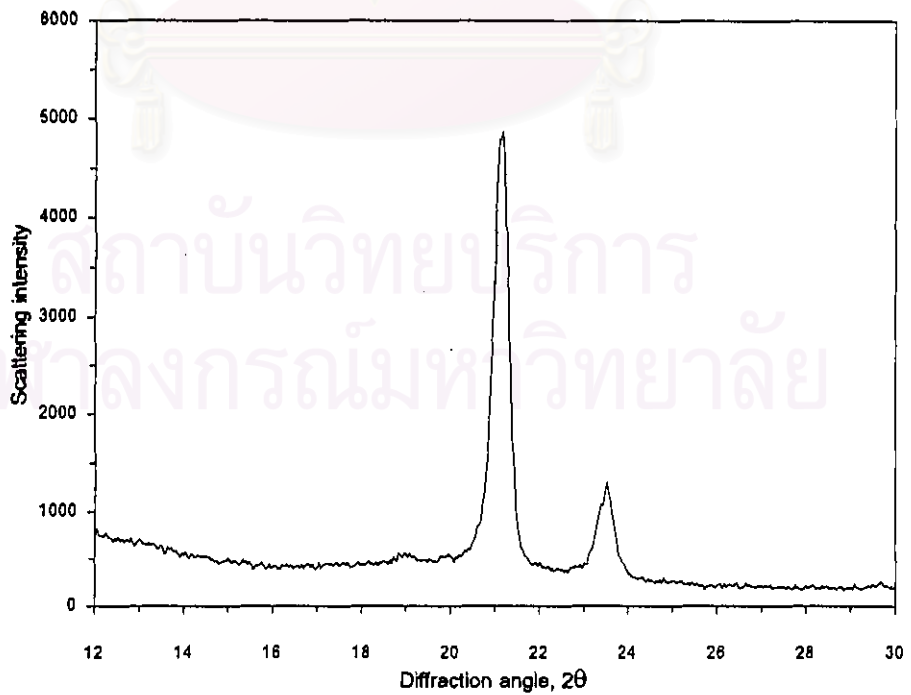


Figure 4.34 X-ray diffraction curve of the recycled HDPE (film).

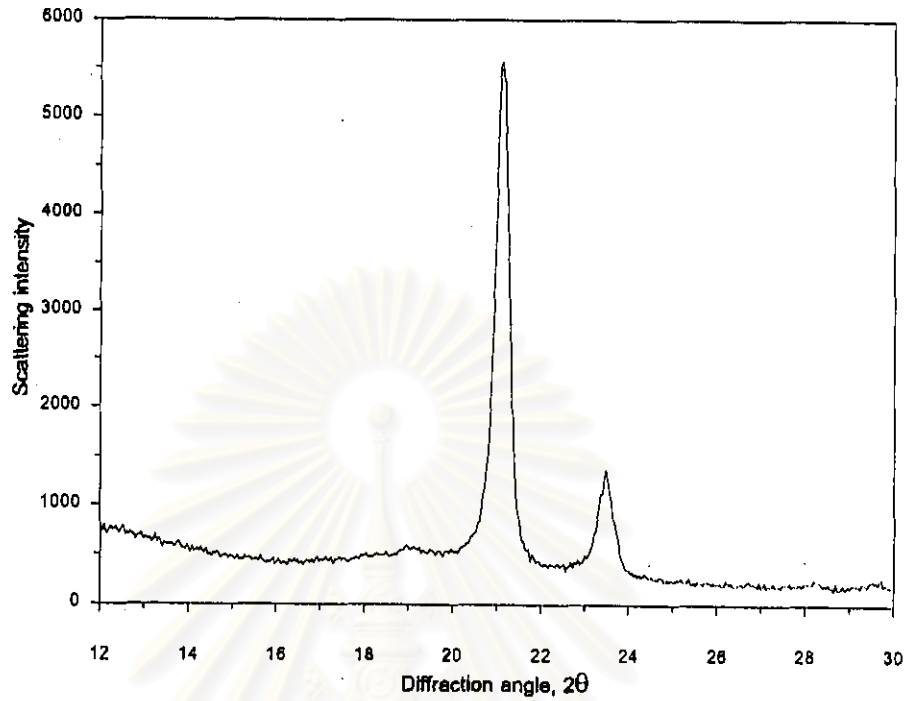


Figure 4.35 X-ray diffraction curve of the recycled HDPE (bottle).

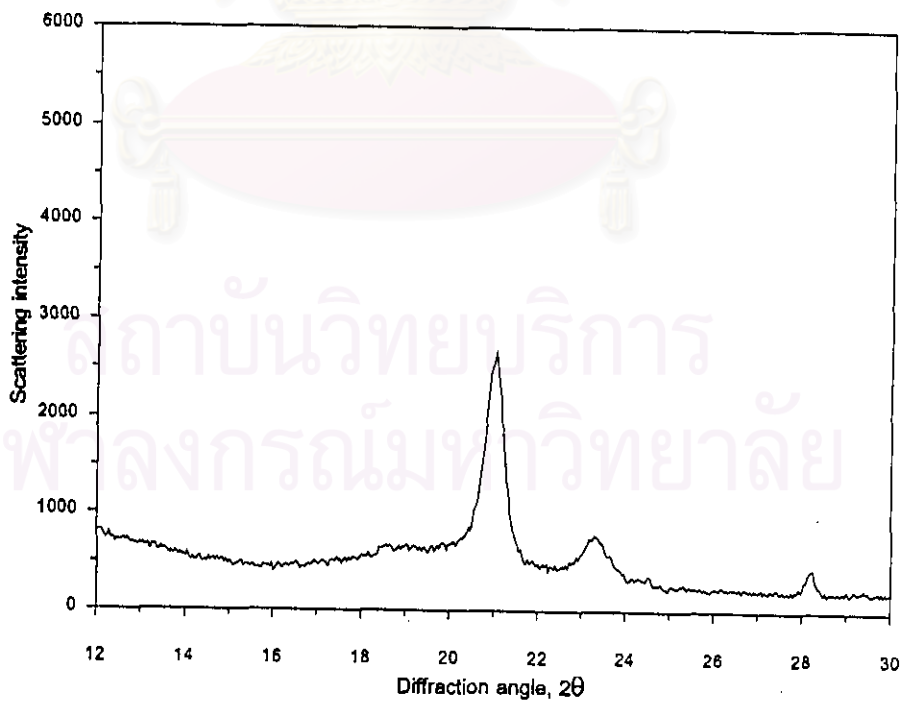
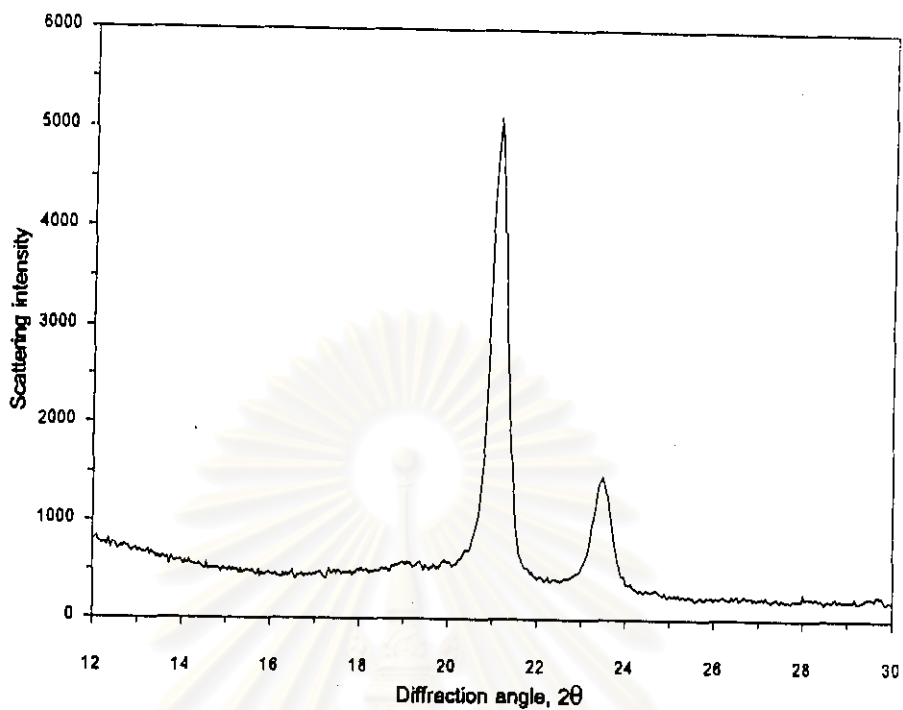
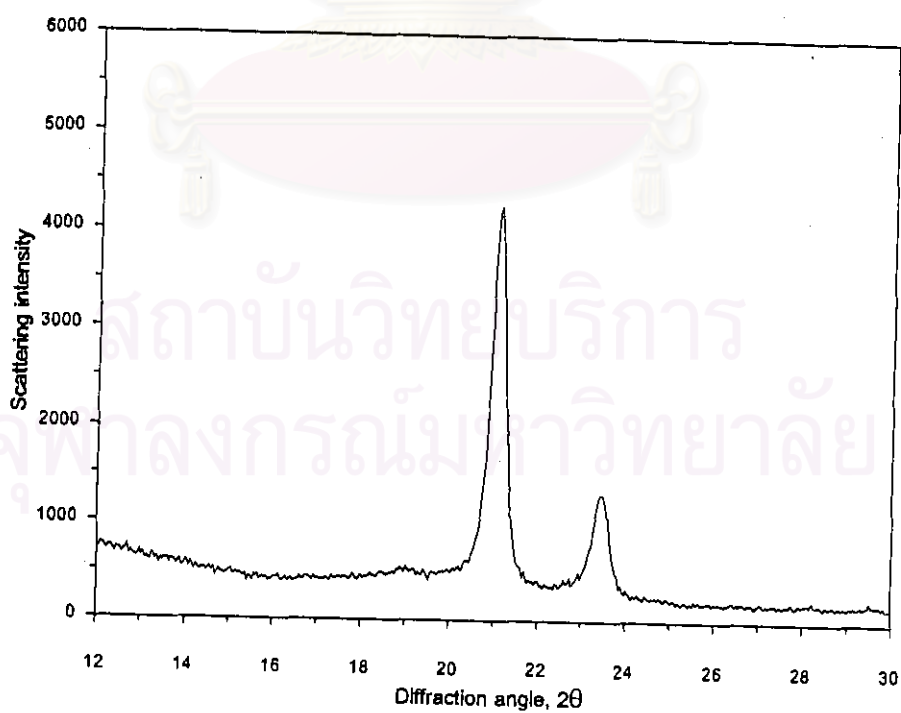


Figure 4.36 X-ray diffraction curve of the recycled LDPE.



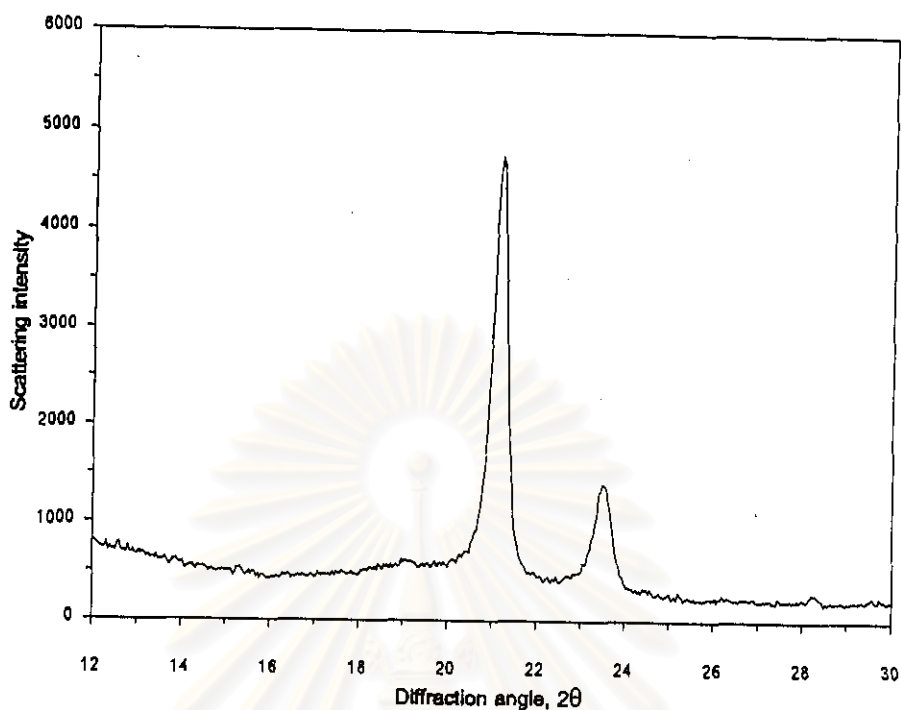
**Figure 4.37** X-ray diffraction curve of the blended PE film.

(virgin HDPE 70%, recycled HDPE (film) 10%, recycled HDPE (bottle)10%, recycled LDPE 10%)



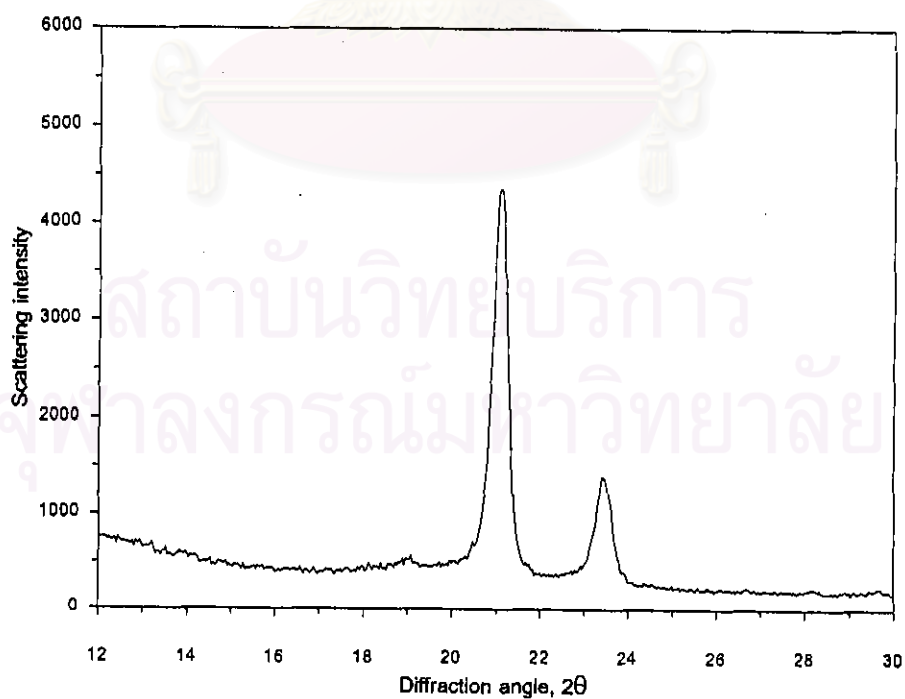
**Figure 4.38** X-ray diffraction curve of the blended PE film.

(virgin HDPE 60%, recycled HDPE (film) 10%, recycled HDPE (bottle)10%, recycled LDPE 20%)



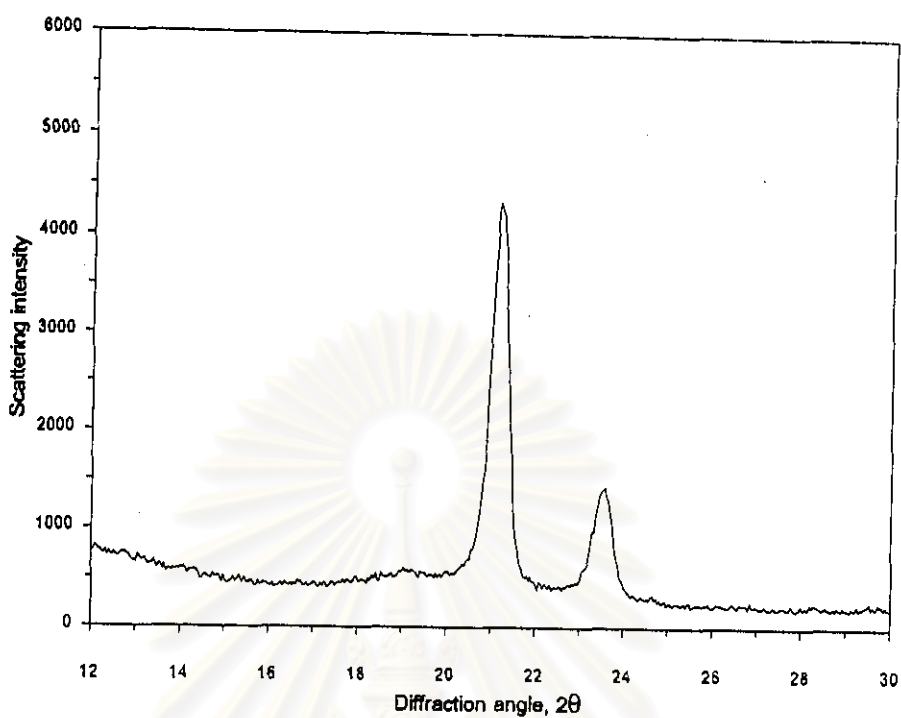
**Figure 4.39** X-ray diffraction curve of the blended PE film.

(virgin HDPE 50%, recycled HDPE (film) 10%, recycled HDPE (bottle) 10%, recycled LDPE 30%)



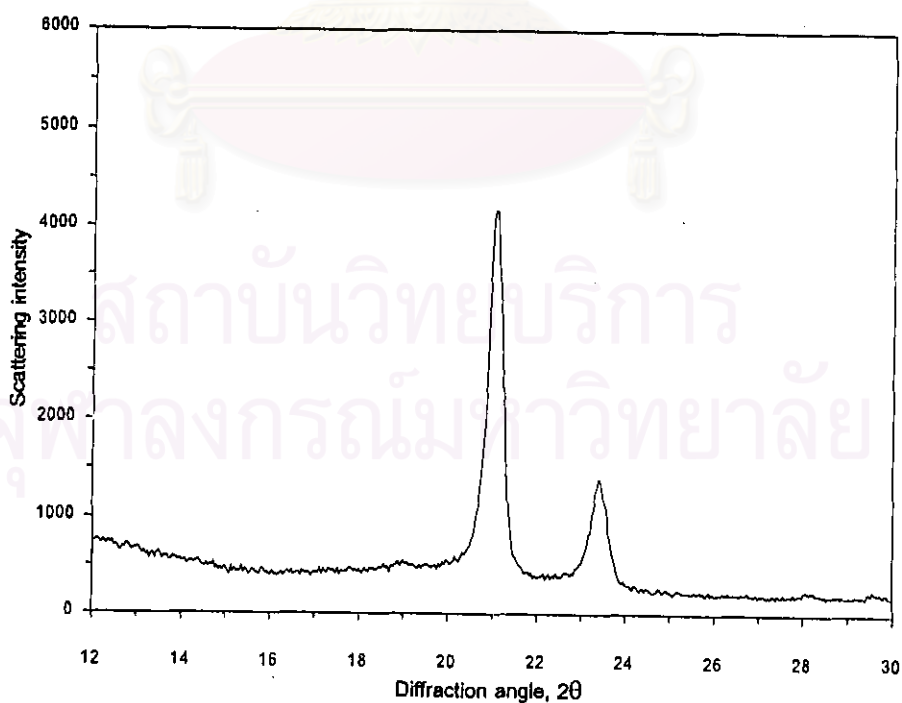
**Figure 4.40** X-ray diffraction curve of the blended PE film.

(virgin HDPE 50%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 10%)



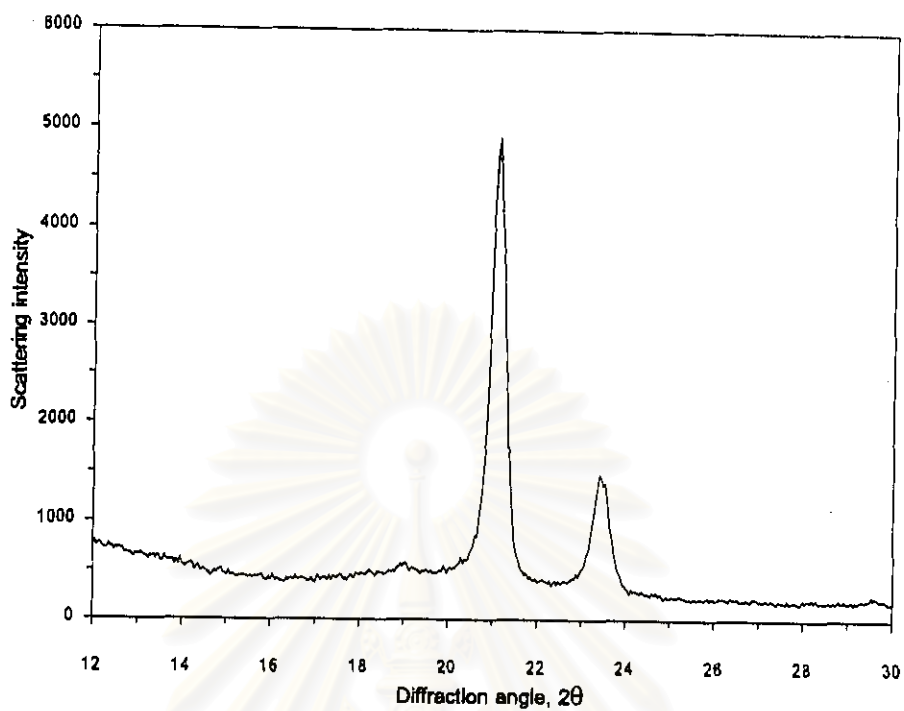
**Figure 4.41** X-ray diffraction curve of the blended PE film.

(virgin HDPE 40%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 20%)



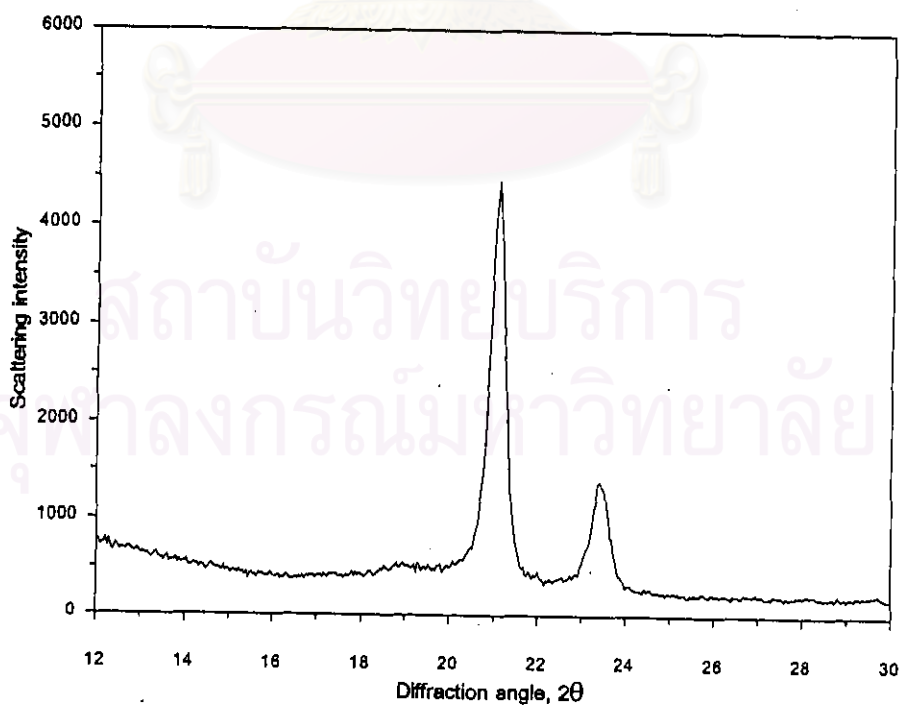
**Figure 4.42** X-ray diffraction curve of the blended PE film.

(virgin HDPE 30%, recycled HDPE (film) 20%, recycled HDPE (bottle) 20%, recycled LDPE 30%)



**Figure 4.43** X-ray diffraction curve of the blended PE film.

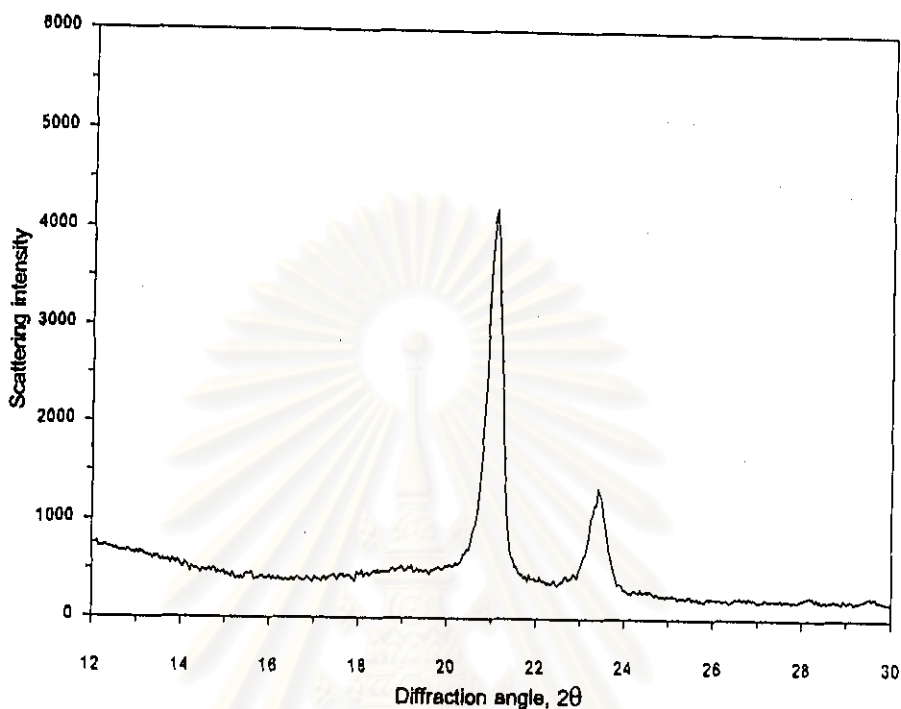
(virgin HDPE 30%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 10%)



**Figure 4.44** X-ray diffraction curve of the blended PE film.

(virgin HDPE 20%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 20%)





**Figure 4.45** X-ray diffraction curve of the blended PE film.

(virgin HDPE 10%, recycled HDPE (film) 30%, recycled HDPE (bottle) 30%, recycled LDPE 30%)

### 4.5.3 Comparison of X-ray Diffraction and DSC.

The result of X-ray diffraction and DSC can now be compared. In Table 4.9, the %crystallinities from X-ray diffraction are compared along side the corresponding heats of fusion from DSC.

**Table 4.9** Comparison of % Crystallinities from X-ray Diffraction and Heats of Fusion from DSC.

| Material (%)   |                            |                              |                  | Crystallinity<br>from<br>XRD<br>(%) | Crystallinity<br>From<br>DSC <sup>a</sup><br>(%) | Heat of Fusion<br>From<br>DSC<br>(J/g) |
|----------------|----------------------------|------------------------------|------------------|-------------------------------------|--|--|
| Virgin<br>HDPE | Recycled<br>HDPE<br>(film) | Recycled<br>HDPE<br>(bottle) | Recycled<br>LDPE |                                     |  |  |
| 100            | -                          | -                            | -                | 77.6                                | 64.3   | 184.7                                  |
| -              | 100                        | -                            | -                | 75.0                                | 62.1   | 178.4                                  |
| -              | -                          | 100                          | -                | 71.7                                | 54.9   | 157.7                                  |
| -              | -                          | -                            | 100              | 51.5                                | 31.7   | 91.1                                   |
| 70             | 10                         | 10                           | 10               | 75.0                                | -  | 183.5                                  |
| 60             | 10                         | 10                           | 20               | 72.1                                | -  | 170.7                                  |
| 50             | 10                         | 10                           | 30               | 70.7                                | -  | 170.5                                  |
| 50             | 20                         | 20                           | 10               | 74.4                                | -  | 174.0                                  |
| 40             | 20                         | 20                           | 20               | 73.8                                | -  | 170.5                                  |
| 30             | 20                         | 20                           | 30               | 70.2                                | -  | 160.4                                  |
| 30             | 30                         | 30                           | 10               | 71.1                                | -  | 163.3                                  |
| 20             | 30                         | 30                           | 20               | 69.5                                | -  | 158.4                                  |
| 10             | 30                         | 30                           | 30               | 67.4                                | -  | 146.1                                  |

<sup>a</sup> the value of % crystallinity of crystalline polyethylene are in the range of 55.3-80.0 % by Calorimetric method [43].

Therefore, both methods (DSC and XRD) in this study used can indicate the crystalline regions in the blended films when compared to the morphology in SEM. It would be expected that, as the content of the recycled LDPE increases, the degree of structural irregularity increases, thus it decreases the ability to crystallize. The presence of short bright fibrils distribution shown in the SEM photographs give the rough surface appearance.

Table 4.10 shows all properties of PE blended with recycled polyethylene resins. The results indicate that the higher proportion of blended recycled LDPE, the poorer the physical and mechanical properties, because the recycled LDPE resins has the greater amount of the lower molecular weight branch chains. Influence of the amorphous region of the recycled LDPE resin over the crystalline region could possibly deform the polymer matrix. The proportion of the recycled LDPE resin affects the morphology and less slightly highly extended chain fibrils, which constitute the back bone. The lamellar stacks of the recycled LDPE film show the network structure of low ordered orientation because the recycled LDPE has low average molecular weights. Therefore, the morphology of the blended film is greatly affected by the recycled LDPE. We can see that the film surface is not smooth with an increased amount of the recycled LDPE leading to a decreased mechanical properties and physical properties. On the other hand, the film is high transparency because the degree of crystallinity is decreased as shown in Table 4.10. The recycled LDPE with high-branching molecular structure and low molecular weights caused the degradation of the PE molecules during cycling processes.

Table 4.11 shows some properties of PE resins based on Thai Industrial Standard (TIS) 816-2538, which was issued by the Ministry of Industry. We choose PE type 3 (HDPE) for comparison with the blended films.

From Table 4.12, we can see the films containing 10-30 percent of recycled LDPE in all blends have the better elongation at break than TIS (27-83 percent). Likewise, the stress at yield of the blends having 10-20 percent of recycled LDPE are better than those of TIS (3-36 percent). Unfortunately, stress at yield of the blended films having 30 percent of recycled LDPE is lower than those from TIS (9-18 percent).

Table 4.10 Morphology and Mechanical Properties of the Blended Recycled Polyethylene Films.

| Material        |                          |                            |                   | Mechanical Properties                 |                                    |            |          |            |          |              |        |              |        | Physical Properties      |                           |            |              |            |                  |
|-----------------|--------------------------|----------------------------|-------------------|---------------------------------------|------------------------------------|------------|----------|------------|----------|--------------|--------|--------------|--------|--------------------------|---------------------------|------------|--------------|------------|------------------|
| Virgin HDPE (%) | Recycled HDPE (film) (%) | Recycled HDPE (bottle) (%) | Recycled LDPE (%) | MFI <sub>2.16/190 °C</sub> (g/10 min) | MFI <sub>5/190 °C</sub> (g/10 min) | $\sigma_y$ |          | $\sigma_b$ |          | $\epsilon_y$ |        | $\epsilon_b$ |        | $E' \times 10^{-3}$ (Pa) | $E'' \times 10^{-4}$ (Pa) | $T_g$ (°C) | Tan $\delta$ | $T_m$ (°C) | $\chi_c$ XRD (%) |
|                 |                          |                            |                   |                                       |                                    | MD (MPa)   | TD (MPa) | MD (MPa)   | TD (MPa) | MD (%)       | TD (%) | MD (%)       | TD (%) |                          |                           |            |              |            |                  |
| 100             | -                        | -                          | -                 | 0.06                                  | 0.27                               | 29.57      | 29.04    | 42.13      | 35.21    | 15.58        | 6.25   | 500          | 664    | 8.0                      | 6.8                       | -119.0     | 0.098        | 131.4      | 77.6             |
| -               | 100                      | -                          | -                 | 0.06                                  | 0.14                               | -          | -        | -          | -        | -            | -      | -            | -      | 7.1                      | 4.7                       | -118.5     | 0.081        | 130.1      | 75.0             |
| -               | -                        | 100                        | -                 | 2.50                                  | 13.22                              | -          | -        | -          | -        | -            | -      | -            | -      | 8.0                      | 4.9                       | -116.7     | 0.104        | 128.1      | 71.7             |
| -               | -                        | -                          | 100               | 4.30                                  | 18.48                              | -          | -        | -          | -        | -            | -      | -            | -      | 4.2                      | 2.2                       | -126.0     | 0.057        | 107.1      | 51.5             |
| 70              | 10                       | 10                         | 10                | 0.07                                  | 0.39                               | 27.10      | 26.35    | 42.94      | 37.36    | 16.27        | 6.02   | 491          | 611    | 6.7                      | 4.6                       | -117.7     | 0.092        | 129.1      | 75.0             |
| 60              | 10                       | 10                         | 20                | 0.15                                  | 0.55                               | 23.65      | 22.99    | 33.28      | 24.85    | 16.67        | 5.54   | 454          | 537    | 6.2                      | 3.3                       | -119.7     | 0.074        | 129.4      | 72.1             |
| 50              | 10                       | 10                         | 30                | 0.17                                  | 0.82                               | 20.65      | 19.72    | 30.27      | 25.81    | 16.17        | 6.16   | 418          | 534    | 4.7                      | 2.7                       | -119.4     | 0.082        | 131.7      | 70.7             |
| 50              | 20                       | 20                         | 10                | 0.16                                  | 0.60                               | 25.67      | 25.10    | 34.56      | 29.57    | 14.46        | 5.94   | 457          | 609    | -                        | -                         | -          | -            | 128.7      | 74.4             |
| 40              | 20                       | 20                         | 20                | 0.18                                  | 0.87                               | 22.50      | 21.36    | 30.02      | 22.50    | 14.41        | 6.32   | 420          | 526    | -                        | -                         | -          | -            | 128.4      | 73.8             |
| 30              | 20                       | 20                         | 30                | 0.28                                  | 1.34                               | 18.49      | 17.36    | 26.42      | 19.84    | 14.00        | 6.59   | 404          | 489    | -                        | -                         | -          | -            | 129.1      | 70.2             |
| 30              | 30                       | 30                         | 10                | 0.18                                  | 0.95                               | 24.16      | 23.89    | 25.88      | 26.66    | 14.35        | 6.61   | 408          | 535    | 6.3                      | 3.9                       | -117.9     | 0.063        | 129.4      | 71.1             |
| 20              | 30                       | 30                         | 20                | 0.30                                  | 1.40                               | 21.20      | 20.51    | 23.46      | 16.46    | 14.28        | 5.52   | 384          | 462    | 6.0                      | 2.8                       | -118.4     | 0.067        | 129.1      | 69.5             |
| 10              | 30                       | 30                         | 30                | 0.47                                  | 2.07                               | 16.85      | 15.39    | 20.98      | 14.92    | 14.70        | 6.44   | 366          | 399    | 5.3                      | 2.7                       | -119.4     | 0.088        | 127.4      | 67.4             |

Table 4.11 Thai Industrial Standard of Polyethylene Resin.

| Item | Properties                               | Specification Control |                 |                |               |         |               |                 |                |               |         |               |                 |                |               |         | Test Method           |
|------|--|-----------------------|-----------------|----------------|---------------|---------|---------------|-----------------|----------------|---------------|---------|---------------|-----------------|----------------|---------------|---------|-----------------------|
|      |  | Kind of Material      |                 |                |               |         |               |                 |                |               |         |               |                 |                |               |         |                       |
|      |  | Type 1                |                 |                |               |         | Type 2        |                 |                |               |         | Type 3        |                 |                |               |         |                       |
|      |  | 1                     | 2               | 3              | 4             | 5       | 1             | 2               | 3              | 4             | 5       | 1             | 2               | 3              | 4             | 5       |                       |
| 1    | Density (g/cm <sup>3</sup> )             | ≤ 0.925               |                 |                |               |         | 0.926-0.938   |                 |                |               |         | ≥ 0.939       |                 |                |               |         | ASTM D 1505           |
| 2    | Melt flow index (g/10 min)               | Less than 0.4         | Over 0.4 to 1.0 | Over 1.0 to 10 | Over 10 to 25 | Over 25 | Less than 0.4 | Over 0.4 to 1.0 | Over 1.0 to 10 | Over 10 to 25 | Over 25 | Less than 0.4 | Over 0.4 to 1.0 | Over 1.0 to 10 | Over 10 to 25 | Over 25 | ASTM D 1238 (2.16 kg) |
| 3    | Stress at yield (MPa)<br>Not less than   | 11.77                 | 11.77           | 8.82           | 7.84          | 5.88    | 13.73         | 13.24           | 11.77          | 11.77         | 9.81    | 19.61         | 19.61           | 14.00          | 13.24         | 11.77   | ASTM D 638            |
| 4    | Elongation at break (%)<br>Not less than | 300                   | 300             | 300            | 100           | -       | 300           | 300             | -              | -             | -       | 300           | 300             | -              | -             | -       | ASTM D 638            |

The data came from the Constitute of Thai Industrial Standard (TIS) 816-2358, the Ministry of Industry, Bangkok, Thailand

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**Table 4.12 Comparison of Thai Industrial Standard of Polyethylene Resin and the Blended Polyethylene Resins.**

| Material        |                          |                            |                   | Mechanical properties                 |                 |          |                   |                 |                     |        |                   |                 | Material Cost<br>(US\$/kg) |
|-----------------|--------------------------|----------------------------|-------------------|---------------------------------------|-----------------|----------|-------------------|-----------------|---------------------|--------|-------------------|-----------------|----------------------------|
| Virgin HDPE (%) | Recycled HDPE (film) (%) | Recycled HDPE (bottle) (%) | Recycled LDPE (%) | MFI <sub>2.16/190 °C</sub> (g/10 min) | Stress at yield |          |                   |                 | Elongation at break |        |                   |                 |                            |
|                 |                          |                            |                   |                                       | MD (Mpa)        | TD (MPa) | % D <sup>a)</sup> | σ <sup>b)</sup> | MD (%)              | TD (%) | % D <sup>a)</sup> | σ <sup>b)</sup> |                            |
| 100             | -                        | -                          | -                 | 0.06                                  | 29.57           | 29.04    | -                 | ± 1.36          | 500                 | 664    | -                 | ± 23.58         | 0.840                      |
| 70              | 10                       | 10                         | 10                | 0.07                                  | 27.10           | 26.35    | +36.26            | ± 0.43          | 491                 | 611    | +83.67            | ± 34.78         | 0.751                      |
| 60              | 10                       | 10                         | 20                | 0.15                                  | 23.65           | 22.99    | +18.92            | ± 1.12          | 454                 | 537    | +65.17            | ± 31.56         | 0.735                      |
| 50              | 10                       | 10                         | 30                | 0.17                                  | 20.65           | 19.72    | +2.91             | ± 0.94          | 418                 | 534    | +58.67            | ± 28.56         | 0.719                      |
| 50              | 20                       | 20                         | 10                | 0.16                                  | 25.67           | 25.10    | +29.42            | ± 0.94          | 457                 | 609    | +77.67            | ± 19.36         | 0.677                      |
| 40              | 20                       | 20                         | 20                | 0.18                                  | 22.50           | 21.36    | +11.83            | ± 1.38          | 420                 | 526    | +57.67            | ± 17.14         | 0.661                      |
| 30              | 20                       | 20                         | 30                | 0.28                                  | 18.49           | 17.36    | -8.62             | ± 0.68          | 404                 | 489    | +48.83            | ± 32.80         | 0.645                      |
| 30              | 30                       | 30                         | 10                | 0.18                                  | 24.16           | 23.89    | +22.49            | ± 1.15          | 408                 | 535    | +57.17            | ± 27.40         | 0.604                      |
| 20              | 30                       | 30                         | 20                | 0.30                                  | 21.20           | 20.51    | +6.37             | ± 1.30          | 384                 | 462    | +41.00            | ± 11.15         | 0.588                      |
| 10              | 30                       | 30                         | 30                | 0.47                                  | 16.85           | 15.39    | -17.80            | ± 1.02          | 384                 | 399    | +27.50            | ± 38.59         | 0.572                      |

1 US\$ = 38.11 Baht (on date 26 Apr 2000 from the Nation).

The cost of PE resin (26 Apr 2000 from Thai Bamroong Import Export Co., Ltd.) is as follows: Virgin HDPE (film) = 0.840 US\$/kg (32 Baht/kg);

Recycled HDPE (film) = 0.525 US\$/kg (20 Baht/kg); Recycled HDPE (bottle) = 0.420 US\$/kg (16 Baht/kg); Recycled LDPE = 0.682 US\$/kg (26 Baht/kg).

<sup>a)</sup> percent deviation calculated for the difference between the sample and value in TIS, <sup>b)</sup> standard deviation

Additionally, we calculated the cost of the blended films. We found that, when the blended films contain more recycled polyethylene resins, the cost is lower from the virgin HDPE of 0.84 US\$ per kg to less than 0.75 US\$ per kg.

Based on Tables 4.10 and 4.12, three blended films containing 70/10/10/10, 50/20/20/10, 30/30/30/10 of virgin HDPE/Recycled HDPE (film)/Recycled HDPE (bottle)/Recycled LDPE are of interest. The possibly highest amount of recycled LDPE is 10%, because the mechanical properties are not too inferior when compared with the type 3 standards. The cost of blended films is also much cheaper (0.751, 0.677, and 0.604 US\$/kg) in comparison the virgin HDPE (0.840 US\$/kg).



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