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<b>Author(s)</b>	<b>Wong, ACY</b>
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# The water permeability of blended polyethylene films

Anthony Chi-Ying Wong

Department of Industrial and Manufacturing Systems Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong

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**Abstract:** Water vapour transmission (WVT) characteristics of blended polyethylene films blown from a 45 mm single-screw extruder were examined. For a given die gap size and blow-up ratio, it was found that WVT basically had an exponential decaying relationship with increasing film thickness. For a given film thickness, molecular orientation on the film in the machine direction could be increased by increasing the size of the die gap. It was found that WVT of blended polyethylene films which involved some proportion of long chain side molecular branches components were more susceptible to die gap size effect than those films only involving linear type molecules. This was thought to be attributed to the more molecular orientation in the machine direction. The results showed that for a given degree of orientation in the transverse direction, WVT decreased if film thickness and die gap size increased. This study also demonstrated that lower WVT required longer saturation time and smaller amount of saturated water vapour obtained.

## 1 Introduction

Considerations given to the selection of polymeric films for packaging are generally based upon the performance requirements of the end-products, processability, cost and availability of the resin in the market. Over the years, polyethylene (PE) film has proved to meet these considerations and has, thus, been a common polymeric packaging material in the industry. The PE film has been used either as a barrier material or as a bonding layer in a multi-layer packaging matrix.

If a linear low density polyethylene (LLDPE) is to be properly processed in a conventional processing machinery originally designed for low density polyethylene (LDPE), modifications on the machine are likely to be required. However, this modification requirement may not be always acceptable since this may involve cost, technical difficulties etc. Apparently, blending LLDPE with other types of PE offers a simple solution to the problem, and such blending practice has already become a common practice in the plastic industry. In fact, blending PE/PE is carried out also for reasons other than those technical ones, such as financial incentives.<sup>1</sup>

Investigations on the subject of PE/PE blends have largely been related to either mechanical, thermal or morphological properties of the blends. A summary in this respect can be found in the work of Wong.<sup>2</sup> The current study, which is part of a large research program on the various aspects of poly-

ethylene film, aims to correlate a selected processing variable with a selected physical property of blended PE films used as a packaging material. To this end, water vapour permeability characteristic was thus selected for the investigation. This barrier behaviour of a packaging material is one of the most essential properties for proper packaging design, no matter if water vapour is to be kept away from or to be maintained in the package.

It is a well known fact that polymeric materials have anisotropic mechanical characteristics. The directional characteristics are sometimes even critical in practical applications. In tubular blown films extrusion, the directional characteristics of the films in the machine direction (MD) are often related by the pulling rate (i.e. film thickness) of the nip roll and the 'drawdownability' of the polymer melt. The characteristics of the films in the transverse direction (TD) are governed by the blow-up-ratio (BUR) which is defined as the ratio of the diameter of the bubble to the diameter of the circular die. In this study, BUR was fixed at 2.5:1 for all the experimental work to enable the directional properties of the films in MD to be isolated and studied. To achieve this, die gap size was chosen as a processing variable for this part of the investigation.

## 2 Experimental

Two families of PE/PE blends were prepared from three pure resins supplied by Exxon Chemical. One

of the families involved LLDPE/LDPE blends, whereas another family was blends of LLDPE/LLDPE. Each family had three blend ratios: 10, 20 and 30%. The density and melt index of the three pure resins and the blends are shown in Table 1 which also includes the sample code.

Sample A was a butene base LLDPE manufactured particularly for film blowing and was therefore chosen as the base blending component. It had a melt index of  $1 \text{ g (10 min)}^{-1}$ . The experience of the resin supplier and that of our own on this resin showed that because of its relatively low flowability (melt index) there were some general difficulties to process this resin in its pure form in a conventional blown film extruder. Bubble instability, melt fracture, etc. were the usual problems associated with pure Sample A during processing. However, blending it with either LDPE or LLDPE of higher melt index were found to be satisfactory to overcome these processing problems. It was therefore on this basis that Sample B and C were chosen as the blending partner with Sample A in this investigation.

Blending was carried out by physically mixing the carefully pre-weighed components before charging the mixture into the hopper of a 45 mm single-screw blown film extruder. The screw had a 25 length-to-diameter ratio, a Maddock mixing device and a barrier flight in its metering zone. It had also a compression ratio of 3.5 to 1. The circular die had a 100 mm diameter and was of a spiral design. The die lip was made detachable for ease of altering the size of the die gap. Three die gap sizes (1, 1.5 and 2 mm) were used in this study. The size of the film bubble was carefully controlled to have a BUR of 2.5 to 1. Cooling of the bubble was provided by a dual orifice cooling air ring. The incoming cooling air was maintained at around 28°C. The frost line height was kept at around 350 mm measured from the die surface, and the stability of the bubble was enhanced by an iris diaphragm. The bubble was carefully controlled to ensure that there was no significant differences in

the bubble configuration from run to run. The screw speed was kept at 50 rpm and care on the process controls was taken to avoid having any melt fracture on the films. Of each blend, five film thicknesses were made. They were: 20, 30, 40, 50 and 60  $\mu\text{m}$ .

Water vapour transmission rate (WVT) was measured by the weight-gain dish method as described in the ASTM E96-93 standard test method. A small amount of anhydrous calcium chloride was placed in a CEAST metal dish which had a 3000  $\text{mm}^2$  exposed area for the test. A small amount of wax was smeared round the mouth of the dish before assembly to make it leak proof. Weighing of the dish to determine the amount of water vapour transmitted was carried out periodically until a constant weight of the dish was detected which took some time. The test samples were kept in a small room, where temperature and relative humidity were controlled at 23°C and 65% respectively during the whole experiment. Each experimental result reported here represented an average of five measurements.

Melt indexes of pure and blended resins were determined by a CEAST Melt Flow Indexer at 2.16 kg and 190°C. The density was measured by a TECHNE's density gradient column.

### 3 Calculations

The amount of water vapour transmitted through the plastic film and absorbed by the anhydrous calcium chloride was determined by the measurement of the weight change,  $G$ . The time,  $t$ , during which  $G$  occurred was also recorded. For each test, when  $G$  per unit area was plotted linearly against  $t$ , a graph similar to Fig. 1 was then obtained. The rate of water vapour transmission was calculated according to eqn (1) as specified by the ASTM, i.e.:

$$WVT = \frac{G}{tA} \quad (1)$$

Table 1. Physical properties of the pure resins and blends

Sample code (pure resin)	Resin	Melt index ( $\text{g 10 min}^{-1}$ )	Density ( $\text{kg m}^{-3}$ )	Application
A	LLDPE	1	918	Blown film
B	LDPE	4	921	Blown film
C	LLDPE	50	926	Injection
Sample code	Blend partners	Blend ratio	Melt index ( $\text{g 10 min}^{-1}$ )	Density ( $\text{kg m}^{-3}$ )
D	A:B	90:10	1.19	918
E	A:B	80:20	1.20	918
F	A:B	70:30	1.22	919
G	A:C	90:10	1.45	919
H	A:C	80:20	2.20	920
I	A:C	70:30	2.66	921

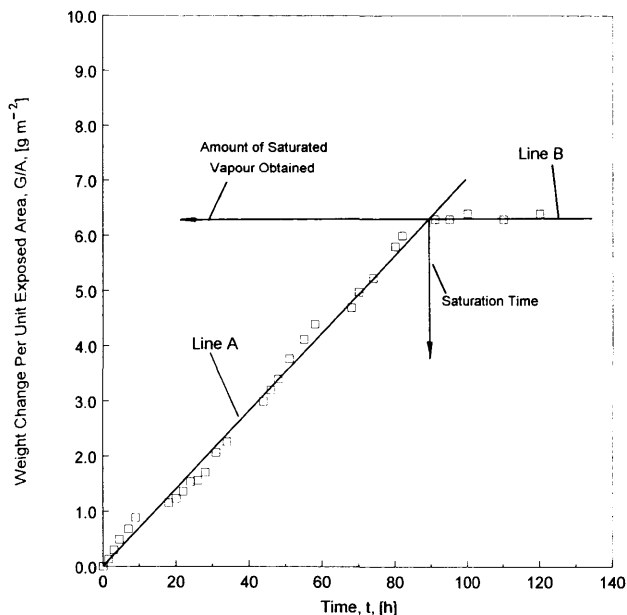


Fig. 1  $G/A$  against  $t$  for blend F (70% LLDPE/30% LDPE), die gap size = 2 mm.

where  $WVT$  is the rate of water vapour transmission,  $g\ h^{-1}\ m^{-2}$ ;  $G$  is the weight change,  $g$ ;  $t$  is the time during which  $G$  occurred,  $h$ , and;  $A$  is the test area,  $m^2$ .

The term,  $G/tA$ , in eqn (1) is apparently the slope (Line A) of the best fit of the first part of the data of the  $G/A$  vs  $t$  plot (i.e. before saturation is reached). It has to be mentioned that the slope must pass through the origin of the graph. The horizontal line (Line B) in Fig. 1 is the saturation point of the transmission. The time beyond which  $G/A$  is basically a constant should be taken as the saturation time. However, none of the experimental data gave a well defined saturation time. Saturation time was therefore determined by taking the intersection between line A and B as shown in Fig. 1.

While  $WVT$  is related to properties of a material, the evaluation of water vapour transmission performance of a material is given by the water vapour permeance which can be calculated from eqn (2):

$$\text{Permeance} = \frac{WVT}{\Delta p} = \frac{WVT}{S(R_1 - R_2)} \quad (2)$$

where  $\Delta p$  is the vapour pressure difference (mm Hg);  $S$  is the saturation vapour pressure at test temperature (mm Hg);  $R_1$  is the relative humidity at the source expressed as a fraction, and;  $R_2$  is the relative humidity at the vapour sink expressed as a fraction.

#### 4 Results and discussions

If a gas or vapour is to pass from one side of a plastic film to the other, there are basically two mechanisms. One of which is that the gas or vapour actually

passes through holes, micro-cracks, etc. of the film, and that the film is then described as porous. The second mechanism involves films without holes or micro-cracks and that the transmission is achieved by means of the process known as permeation. Thin organic films can be porous as well as permeable. The degree of porosity in thick films may be less than that of the thin films. However, the overall permeation of thicker films cannot be eliminated simply by increasing film thickness though the rate of their permeation may be reduced.

The mechanism of gas or vapour transmitted through a piece of film can be complicated, and there are many factors governing the rate of transmission. In the simplest case for engineers and applied scientists, it may be adequate for us to realize the difference in pressure exerted by the gas or vapour on each face of the film at constant temperature is directly proportional to the amount of gas or vapour passing through the film of unit exposed area and thickness; and that the thicker the film, the less the quantity of gas or vapour may pass through. An excellent comprehensive discussion on this subject can be found from the work of Hennessy *et al.*<sup>3</sup> Their analysis and discussions covered a fairly broad scope of the various effects on permeability, including nature of the polymers, temperature and pressure effects etc. Another comprehensive piece of work on permeation properties of polymeric material was given by Naylor.<sup>4</sup> On the investigation of the mechanical properties and water vapour permeability of polypropylene (PP) and poly(ethylene terephthalate) (PET) blends which were used as geotextile materials, Bataille *et al.*<sup>5</sup> noted that water vapour permeation was a function of the blend ratio. They reported that the experimental data were always smaller than those estimated. Bataille *et al.* also suggested that the composition of PP present in the blend was of the controlling factor in the overall permeability. Subramanian and Mehra<sup>6</sup> modified the Barrer *et al.*<sup>7</sup> expression on permeability as follows:

$$\frac{1}{P_c} = \frac{\phi_1}{P_1} = \frac{\phi_2}{P_2} \quad (3)$$

where  $P_c$  is the permeability of the composite;  $P_1$ ,  $P_2$  is the permeability of the individual components;  $\phi_1$ ,  $\phi_2$  is the volume fraction of the two components.

From their work on PE/nylon blends, Subramanian and Mehra were able to conclude that the overall barrier property of the film could be enhanced if the blend was what they termed 'laminar blend'. Similar to Subramanian and Mehra, Blahovici and Brown<sup>8</sup> also studied permeation from morphological and micro-structure point of view. Their experimental results revealed that the increase in oxygen permeation of a polymer blend was closely related to the layered structure of the blend. Temperature

effects on permeation were recently investigated by Tokatli and Ozilgen,<sup>9</sup> however, their work mainly dealt with mono-layer of LDPE. Despite most of this work having enhanced our knowledge on the permeability of plastic films, none of them has attempted to relate this property of a polymeric film to molecular orientation and other processing variables which are the focus of this work.

Since the resin densities of all the blends shown in Table 1 were virtually constant and the difference in the melt index was not a substantial percentage, *WVT* was therefore assumed to be independent of these two resin properties throughout this work. All the experimental data obtained in this study were firstly plotted in the same manner as Fig. 1 (i.e. *G/A* vs *t*) for the basic analysis on *WVT*. Only one set of the results is presented in Fig. 1, which is meant to illustrate the shape of the curve. The gradients of the first part of the curves (i.e. Line A of Fig. 1) were obtained with good confidence. As mentioned above, such slope corresponded to rate of transmission (*WVT*). Transferring this rate of transmission for the study of thickness effect, Figs 2-7 were therefore constructed. The graphs presented here have the same scale on their *x* and *y* axis for convenient comparison. Of all the five film thicknesses tested, it can be seen from the graphs that *WVT* was a function of the film thickness as expected. However, the decrease of *WVT* with increasing film thickness does not take a linear relationship. Rather, the results appeared to be best described by exponential decaying characteristics. Although Hennessy *et al.*<sup>3</sup> had also noted that *WVT* decreased with increasing film thickness, they had not reported the

type of the decreasing characteristics between these two parameters.

On the effect of blend ratio, the results clearly showed that, for a given film thickness and die gap size, *WVT* increased with increasing LDPE proportion in the blends of LLDPE/LDPE, whereas *WVT* of LLDPE/LLDPE blends appeared to be insignificantly affected by the blend ratio. Regardless of the type and proportion of the blends, if one would superimpose all the graphs one over each other, it can be seen that there is a general trend on the effect

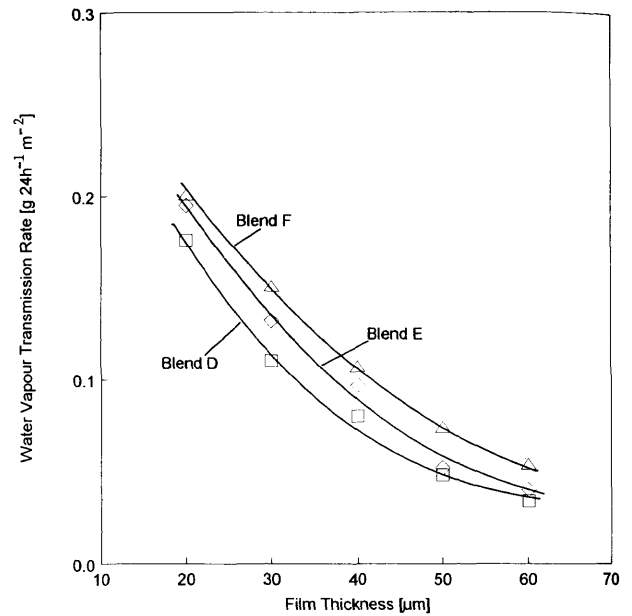


Fig. 3 Relationship between *WVT* and film thickness (LLDPE/LDPE blends, die gap size = 1.5 mm).

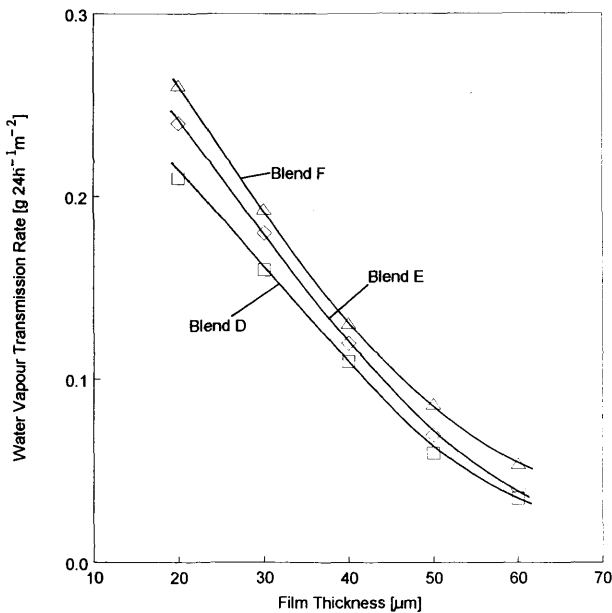


Fig. 2 Relationship between *WVT* and film thickness (LLDPE/LDPE blends, die gap size = 1 mm).

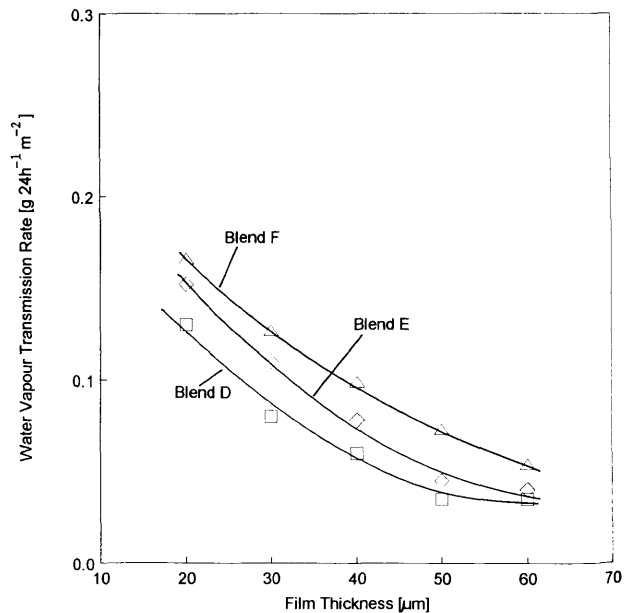


Fig. 4 Relationship between *WVT* and film thickness (LLDPE/LDPE blends, die gap size = 2.0 mm).

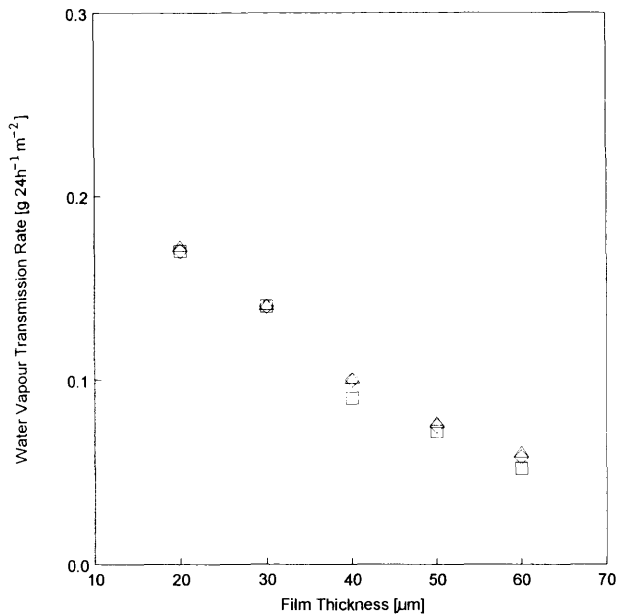


Fig. 5 Relationship between *WVT* and film thickness (LLDPE/LLDPE blends, die gap size = 1.0 mm) □ Blend G, ◇ Blend H, △ Blend I.

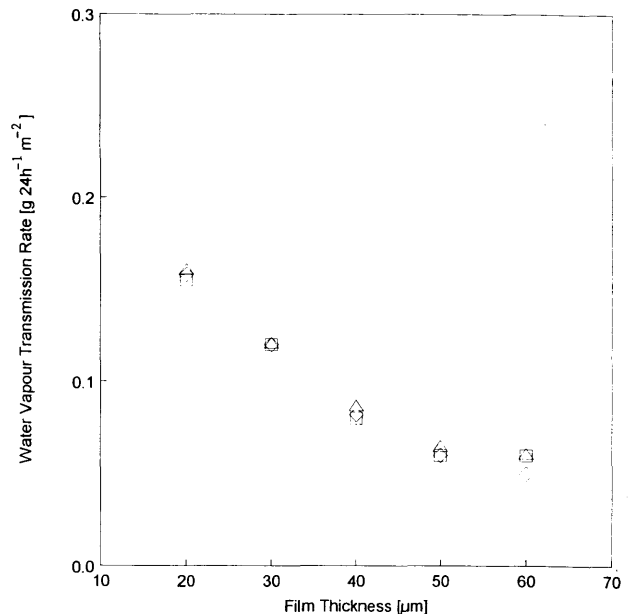


Fig. 7 Relationship between *WVT* and film thickness (LLDPE/LLDPE blends, die gap size = 2.0 mm) □ Blend G, ◇ Blend H, △ Blend I.

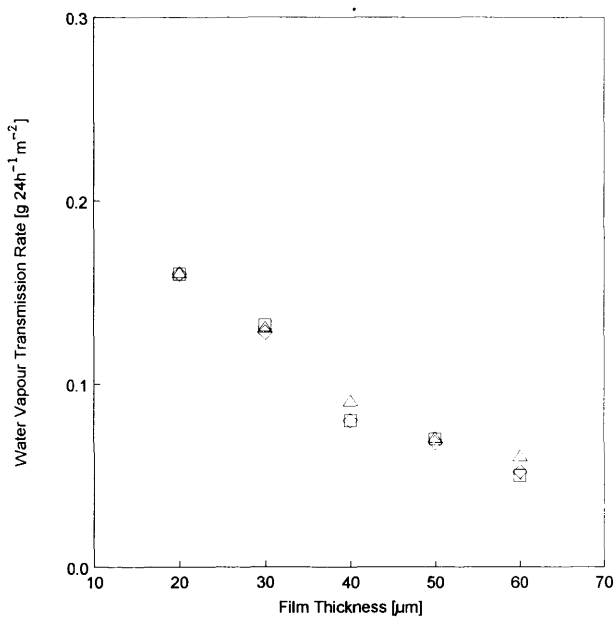


Fig. 6 Relationship between *WVT* and film thickness (LLDPE/LLDPE blends, die gap size = 1.5 mm) □ Blend G, ◇ Blend H, △ Blend I.

of the die gap size. All the experimental data demonstrated that *WVT* was apparently decreased with increasing die gap size (Figs 2–7). Similar to film thickness and blend ratio effects, the die gap size effects on *WVT* of LLDPE/LLDPE was again to a much lesser extent than those on LLDPE/LDPE blends.

It is a well-known fact that most of the performance of extruded polymer products are related to the alignment of their molecular structure and that the

molecular structure (or orientation) is, in turn, highly dependent on the processing variables and conditions. The extent of molecular orientation in the machine direction (MD) of an extruded plastic blown film will depend on the rate of pulling of the bubble by the nip roll located at the top of the collapsing tower of a blown film extrusion line. The size of the bubble during an extrusion process, on the other hand, will determine the molecular orientation in the transverse direction (TD). Bubble size is generally expressed in terms of blow-up-ratio (BUR) which is defined as the diameter of the bubble to the diameter of the die. The reasons for the selection of the types of resins for this work largely was for the investigation of molecular orientation effect on *WVT*. Since LDPE has many more long chain side branches in its molecules than the LLDPE, it was believed that if there was any difference in the performance of *WVT*, part of the reason could be attributed to this intrinsic difference in the basic molecular structure between the types of resins. The BUR for this investigation was fixed at 2.5:1. With this process setting, all blends would then have the same extent of induced molecular orientation in TD, leaving MD to be the variable for the study of molecular orientation effect. In this respect, films of five different thicknesses were blown. For a given screw speed, BUR and die gap size, the thinner the film, the more orientation in the polymer molecules will be induced. The extent of orientation can be reflected from the drawdown ratio which is defined as the ratio of the size of a die gap to the ultimate thickness of a film. It follows that, for a given film thickness, larger molecular orientation will be induced

**Table 2. Drawdown ratio of the film**

Die gap size (mm)	Drawdown ratio				
	20	30	40	50	60
1.0	50.0	33.3	25.0	20.0	16.7
1.5	75.0	50.0	37.5	30.0	25.0
2.0	100.0	66.7	50.0	40.0	33.3

from a bigger drawdown ratio. Table 2 summarizes the relationship of the drawdown ratio between the film thickness and die gap size. It has to be stressed that the calculation of the drawdown ratio presented in Table 2 was purely based on the definition. Die swell was not taken into account. With the help of this definition, it is clear that, in this study, the molecules were more aligned and oriented in MD for thinner films than for thicker films.

Careful analysis of the results of the LLDPE/LDPE blends shown in Figs 2-4 reveals that for a given blend ratio, *WVT* decreased with increasing die gap size; and that for a given die gap size, *WVT* decreased with decreasing blend ratio of LDPE. It has already been mentioned that for a given film thickness, large die gap size will induce more molecular orientation than small die gap size. Molecules will then be more aligned in MD giving rise to a larger resistance for the water vapour molecules to diffuse through, and hence, a decrease in *WVT*. Indeed, high density polyethylene (HDPE) is known to have a much better vapour transmission resistance than other polyethylene. For the blends of LLDPE/LDPE, no significant molecular orientation was found to have been induced by either increasing die gap size or drawdown ratio on the *WVT* was found. The *WVT* of the LLDPE/LDPE blends was therefore only slightly affected, if at all, by the die gap size. This was reflected in Figs 5-7. For the purpose of illustrating the difference in the effect of die gap size just discussed, selected *WVT* data were extracted from both families of the blends and shown in Figs 8 and 9.

When comparing the *WVT* between the two types of blends studied, the results clearly demonstrated that for thin films (say  $\leq 30 \mu\text{m}$ ), LLDPE/LDPE had a faster *WVT* than that of LLDPE/LLDPE. This again can be explained in terms of the presence of the long chain side molecular branches in the LLDPE/LDPE blends. The presence of the long chain side branches have in fact created more free volume for vapour to transmit through, resulting a higher *WVT*. On the contrary, there is no marked difference in the *WVT* of the thicker films (say above  $50 \mu\text{m}$ ) between the two types of blends, at least within the experimental conditions studied in this work. However, it is envisaged that the *WVT* of thicker LLDPE/LDPE films can be increased if there exists a bigger driving

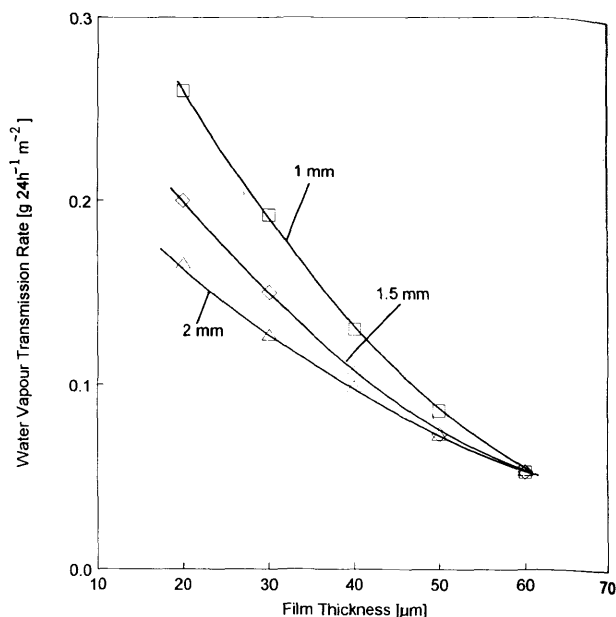


Fig. 8 Die gap size effect on *WVT* (LLDPE/LDPE blends).

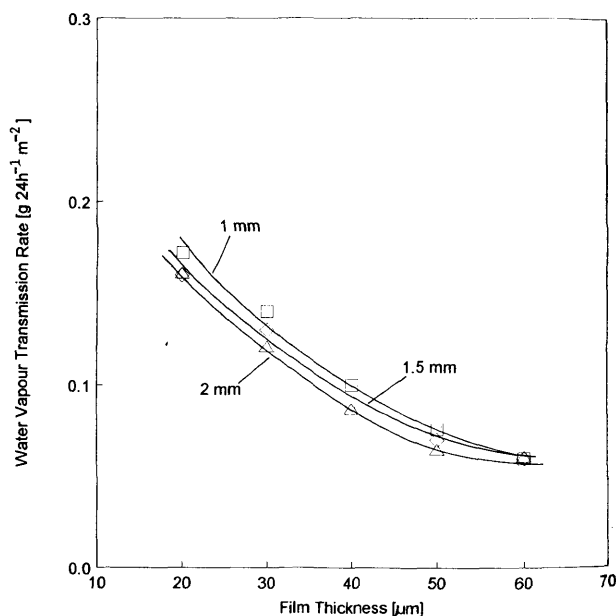


Fig. 9 Die gap size effect on *WVT* (LLDPE/LLDPE blends).

force of permeation between the two surfaces of the film, e.g. larger pressure drop.

The saturation time ( $t_{\text{sat}}$ ) and amount of saturated water vapour ( $m_{\text{sat}}$ ) obtained from the graphical method mentioned above for all the tests carried out in this study are reported in Table 3. It can be seen that, generally, the lower the *WVT*, the higher the  $t_{\text{sat}}$  but the smaller the  $m_{\text{sat}}$ . This indicates that low *WVT* materials will require a longer time to reach saturation and the amount of vapour saturated will be smaller than those of high *WVT* materials.

**Table 3. Saturation time and amount of water vapour saturated of all the samples tested**

Die gap size (mm)	Blend	Blend ratio (%)	Film thickness ( $\mu\text{m}$ )									
			20		30		40		50		60	
			$t_{\text{sat}}$ (h)	$m_{\text{sat}}$ ( $\text{g m}^{-2}$ )	$t_{\text{sat}}$ (h)	$m_{\text{sat}}$ ( $\text{g m}^{-2}$ )	$t_{\text{sat}}$ (h)	$m_{\text{sat}}$ ( $\text{g m}^{-2}$ )	$t_{\text{sat}}$ (h)	$m_{\text{sat}}$ ( $\text{g m}^{-2}$ )	$t_{\text{sat}}$ (h)	$m_{\text{sat}}$ ( $\text{g m}^{-2}$ )
1.0	A:B	90:10	74	15.5	77	12.3	83	9.1	90	5.4	102	3.6
		80:20	72	17.3	76	13.7	83	10.0	89	6.2	100	4.0
		70:30	66	17.2	75	14.4	82	10.7	85	7.3	90	4.8
1.5	A:B	90:10	81	14.3	85	9.4	94	7.5	100	4.8	102	3.5
		80:20	78	15.2	82	10.8	84	8.1	92	4.8	100	4.0
		70:30	75	15.0	78	11.7	85	9.0	88	6.4	91	4.8
2.0	A:B	90:10	82	10.7	90	7.2	96	5.8	102	3.6	103	3.6
		80:20	80	12.2	86	9.5	90	7.0	100	4.5	100	4.0
		70:30	77	12.7	82	10.3	84	8.2	87	6.3	91	4.8
1.0	A:C	90:10	78	13.3	80	11.2	87	7.8	95	6.8	102	5.3
		80:20	77	13.1	81	11.3	86	8.6	96	7.2	103	6.0
		70:30	78	13.4	80	11.2	86	8.6	95	7.2	100	6.0
1.5	A:C	90:10	81	13.0	86	11.4	91	7.3	95	6.7	103	5.2
		80:20	82	13.1	87	11.1	92	7.4	96	6.5	102	5.3
		70:30	81	13.0	86	11.2	91	8.2	95	6.7	100	6.0
2.0	A:C	90:10	82	12.7	86	10.3	90	7.2	100	6.0	100	6.0
		80:20	80	12.6	86	10.3	90	7.4	100	6.0	102	5.1
		70:30	81	13.0	86	10.3	92	7.9	100	6.4	100	6.0

**Table 4. Permeance of all the samples tested**

Die gap size (mm)	Blend	Blend ratio (%)	Permeance ( $\times 10^{-9} \text{ g Pa}^{-1} \text{ s}^{-1} \text{ m}^{-2}$ )				
			Film thickness ( $\mu\text{m}$ )				
			20	30	40	50	60
1.0	A:B	90:10	1.30	1.00	0.70	0.38	0.22
		80:20	1.50	1.10	0.76	0.45	0.25
		70:30	1.70	1.20	0.83	0.55	0.34
1.5	A:B	90:10	1.10	0.70	0.51	0.30	0.22
		80:20	1.20	0.84	0.62	0.33	0.25
		70:30	1.30	0.95	0.68	0.46	0.34
2.0	A:B	90:10	0.83	0.51	0.38	0.22	0.22
		80:20	0.97	0.70	0.50	0.29	0.25
		70:30	1.10	0.80	0.63	0.46	0.34
1.0	A:C	90:10	1.10	0.89	0.57	0.46	0.33
		80:20	1.10	0.89	0.64	0.48	0.37
		70:30	1.10	0.89	0.64	0.48	0.38
1.5	A:C	90:10	1.00	0.84	0.51	0.45	0.32
		80:20	1.00	0.81	0.51	0.43	0.33
		70:30	1.0	0.83	0.57	0.45	0.38
2.0	A:C	90:10	0.99	0.76	0.51	0.38	0.38
		80:20	1.00	0.76	0.52	0.38	0.32
		70:30	1.00	0.76	0.55	0.41	0.38

The performance of the water vapour transmission of all the samples tested in this work was evaluated by the term known as permeance. The results are reported in Table 4. The calculation was based on eqn (2) and, by taking the saturation vapour pressure ( $S$ ) at the test temperature as 21 mm Hg,<sup>10</sup> the relative humidity at the source ( $R_1$ ) as 0.65 (i.e. 65%) and the relative humidity at the vapour sink ( $R_2$ ) as 0 (i.e. 0%). From the definition of eqn (2), it is obvious

that the permeance was also affected by film thickness, die gap size and molecular orientation in the same trend and manner as  $WVT$ .

## 5 Conclusions

For a given film thickness and blow-up ratio, it was found that  $WVT$  of blended PEs involving some proportion of long chain side branches molecules



such as LDPE was more susceptible to die gap size effect. This was believed to be attributed to the fact that, during processing, the long chain side branches molecules were made to orientate in the direction of pulling. The consequence of this orientation was that the molecules then became more linear and, hence, the free volume was reduced and the overall resistance to vapour transmission was increased. This analysis was supported by the experimental results of LLDPE/LLDPE blends. The molecular structure of the components of LLDPE/LLDPE blends were linear. Employing a bigger die gap size to increase molecular orientation was found to have little effect on the *WVT* of films made from those blends. A general trend was also observed that lower water vapour transmission rate would have a longer saturation time but a smaller amount of saturated water vapour was obtained.

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