



# Oxygen barrier properties of biaxially oriented polypropylene/polyvinyl alcohol blend films

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## Abstract

The biaxially oriented PP/PVA blend film was prepared and had the higher oxygen barrier property by about 130 times than that of a biaxially oriented PP film. When the viscosity ratio ( $\eta_{d(PVA)}/\eta_{m(PP)}$ ) decreased, the dispersed PVA phase was developed into platelets during stretching process. Oxygen permeability was dependent on the number and size of PVA platelet. However, the Oxygen permeability was not sensitively changed in above 25 wt% of PVA. To obtain excellent barrier property, the optimum amount of plasticizer and initiator was required. A pasticizer was related to the size and degree of crystallization of PVA platelet. An initiator played the role of a compatibilizer. The oxygen barrier was enhanced with increasing the viscosity of PP and draw ratio. The higher viscosity of PP was advantageous for preventing the delamination of a blend film, and the moisture vapor permeability was not affected with the laminar structure. As a result, the biaxially oriented PP/PVA blend film had the potential of substituting for the PVDC coated BOPP film.

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*Keywords:* Polymer blend; Oxygen permeability; Laminar morphology

## 1. Introduction

The biaxially oriented polypropylene (BOPP) film has widely used for food packaging, but does not have oxygen barrier property. The barrier BOPP film is prepared to coat with a barrier coating material such as a PVDC. However, PVDC has produced dioxin when burning-out it at the low temperature.

Beside the coating method, the blending method of a barrier material and a polyolefin has big attention due to the low manufacturing cost [1–6,14]. The Dupont (USA) had developed HDPE/Nylon blend for agricultural chemical or organic solvent container and fuel tank [4,6]. In addition, the attempts for developing LDPE/EVOH and PET/EVOH blend film for food packaging film have continued [7–10]. However, most of barrier films and bottles have prepared through co-extrusion, and the manufacturing cost of this film is high due to the high investment and difficult process control [1].

Therefore, it is necessary to study novel polymer blend film in order to substitute for the PVDC coated BOPP film. In general, the PVA has the excellent gas barrier property and low price [3]. In this study, PP/PVA mixture was melt-blended through a twin screw extruder and then stretched biaxially in order to obtain the wider and thinner platelets. The morphology and oxygen permeability of PP/PVA blend film had evaluated as a function of the amount or viscosity of component respectively, and draw ratio.

## 2. Experimental

### 2.1. Materials

Four commercial grades of homo-polypropylene (PP) were obtained from the Hyosung corporation (Korea) as pellet form: PP1 (M.I. (g/10 min, 230 °C, 2160 g) = 2.5, density (g/mL) = 0.90), PP2 (M.I. = 5.5, density = 0.90), PP3 (M.I. = 8.0, density = 0.90), PP4 (M.I. = 10.0, density = 0.90). One commercial grade of polyvinyl alcohol (PVA) was supplied by Kuraray Co. (Japan) in

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granule form: Poval PVA-205 (hydrolysis (mol%) = 68.5–89.5, viscosity (mPa·S, 4% aqueous solution, 20 °C) = 4.6–5.4). The glycerin (purity >98.5%) of the Sakamoto Yakuhin Kogyo Co. (Japan) was used as a plasticizer. Perkadox 14 (purity = 40%) was obtained from the Akzo nobel chemicals Inc. (Netherlands), and the maleic anhydride (M.A.) was supplied by NOF corporation (Japan).

## 2.2. Blend preparation

PVA and glycerin were mixed in a super mixer at 1000 rpm for 3 min and then dried in a dehumidifying dryer at 90 °C for 3 h. An initiator and 1 phr of M.A. were mixed with 100 phr of PP in a super mixer, and then the dried PVA was added at 1000 rpm for 3 min. The PP/PVA mixture reacted in a twin screw extruder ( $\Phi = 65$  mm,  $L/D = 30.5$ , Ikegai (Japan): Model PCM 65-30.5-3N) and the PP/PVA master batch (M/B) was prepared from the die-face cutter (Sewon ENG (Korea): Model ES-40P). At this time, the temperature distributions of a twin screw extruder were 140, 180, 205, 210 and 200 °C, and the screw speed was 200 rpm. In order to obtain a 3-layer casting sheet, the PP/PVA M/B was inputted into the main single extruder (Shinwha machine (Korea):  $\Phi = 65$  mm,  $L/D = 29$ ), and the pure PP resin was inputted into two side single extruders ( $\Phi = 30$  mm,  $L/D = 29$ ), and then extruded from a 3-layer slit die. The temperatures of main extruder were set to 180, 190, 200 and 210 °C, and the die was 210 °C. The screw speed had kept 50 rpm. In order to prevent orientation into the machine direction (MD), the ratio between the extrusion speed and the winding speed by a cooling roll was fixed at 1. The thickness ratio between the core PP/PVA blend layer and the both skin PP layers was 5/90/5%. The casting sheet was cut in the size of 100 mm  $\times$  100 mm, and oriented into MD at the speed of 1000 mm/min using the lab. stretcher (Toyoseki Co. (Japan): model B ( $\times 10$ )) and then oriented into the transverse direction (TD) at 155 °C. The prepared 3-layer film has two merits compared with a single PP/PVA film. First, the stability to moisture was largely enhanced. PVA is very weak to moisture, and two PP skin layers protected the PP/PVA blend layer from moisture. Second, the degree of transparency was improved. A single PP/PVA blend film has a high haze value due to the roughness of surface. Two PP skin layers reduced the roughness of surface and the haze value of 3-layer film decreased drastically from 29–31% to 2–3%.

In BOPP mass production line (Toshiba Co. (Japan)), the casting sheet was oriented by 4.5 times into MD by means of the difference of roll (temperature: 120–130 °C) speed, and then by 8 times into TD in inner of the Ten Tor tunnel (temperature: about 150 °C). The take-off speed was 70–80 m/min.

## 2.3. Viscosity measurement

The shear viscosity was measured according to shear rate

using the Instron Capillary Rheometer, Model 3211. The used capillary nozzle was No. 1708 (diameter: 0.0502 in., length: 2.0058 in.).

The Melt flow Index (M.I.) was measured using the model 522 melt flow indexer of Toyoseiki company according to ASTM D1238.

## 2.4. Morphological analysis

In order to analyze the morphology of the extruded blend film, blend samples were fractured in liquid nitrogen and the fracture surfaces were sputter coated with gold-palladium alloy. The morphology was observed using a scanning electron microscope (SEM)—Hitachi (Japan) model S-500C.

## 2.5. Oxygen permeability measurement

The oxygen permeability of a blend film was measured using the procedure and instrument described in ASTM D3985. The model OX-TRAN 100A of the Mocon corporation (USA) was used and the diffusion cell area was 100 cm<sup>2</sup>.

## 2.6. Moisture vapor permeability measurement

The moisture vapor permeability was measured using the model PERMATRAN-W1A of the Mocon corporation (USA) according to ASTM F-1249.

## 2.7. Delamination test

The interfacial adhesive between skin and core layers of a blend film was measured using the universal test machine (Instron Model 6021). An adhesive tape (25 mm of width) was attached to one surface of the film, and then a force was quickly applied to the tape, the skin layer was delaminated from the core layer. Thereafter, the delaminated skin layer was held by the UTM clip and pulled at the speed of 300 mm/min.

## 2.8. Thermal measurement

Thermal analysis was performed from 30 to 250 °C at a rate 20 °C/min under nitrogen atmosphere by the model TA-7 DSC of Perkin–Elmer company.

## 2.9. FT-IR analysis

In order to check whether the reaction occurred or not between PP resin and PVA, the PP/PVA blend film was analyzed using the Digilab company (USA): model FTS-60. The pure PVA film was prepared in such a manner that the 5% PVA aqueous solution was cast on petri-dish and then dried in vacuum oven.

### 3. Result and discussion

FT-IR analysis was performed to confirm whether the reaction of PP resin and PVA occurred during melt blending or not. Fig. 1(a)–(c) showed the FT-IR spectra of the partially hydrolyzed PVA (87 mol%) film, BOPP film and PP/PVA blend film. Fig. 1(d) displayed a FT-IR spectrum representing the subtraction of the spectra of Fig. 1(a) and (b) from that of Fig. 1(c). In Fig. 1(d), new ester peak appeared at  $1736\text{ cm}^{-1}$ , which was attributed to graft copolymer between the PVA and the PP resin, and it means that new copolymer was formed during melt blending.

#### 3.1. Rheological property

Fig. 2 represented the shear viscosity of PP resin and PP/PVA blend according to shear rate at 190 and 230 °C. The Poval PVA-205 impregnated with 30 phr of glycerin was flown down from capillary die, even when any shear stress was not applied. Therefore, the shear viscosity of Poval PVA-205 was not measured. The viscosity of PP resin was larger than that of PP/PVA blend at the low shear rate, and the viscosity difference between PP and PP/PVA blend became larger with increasing the measuring temperature. This result means that the shear viscosity of PVA is lower

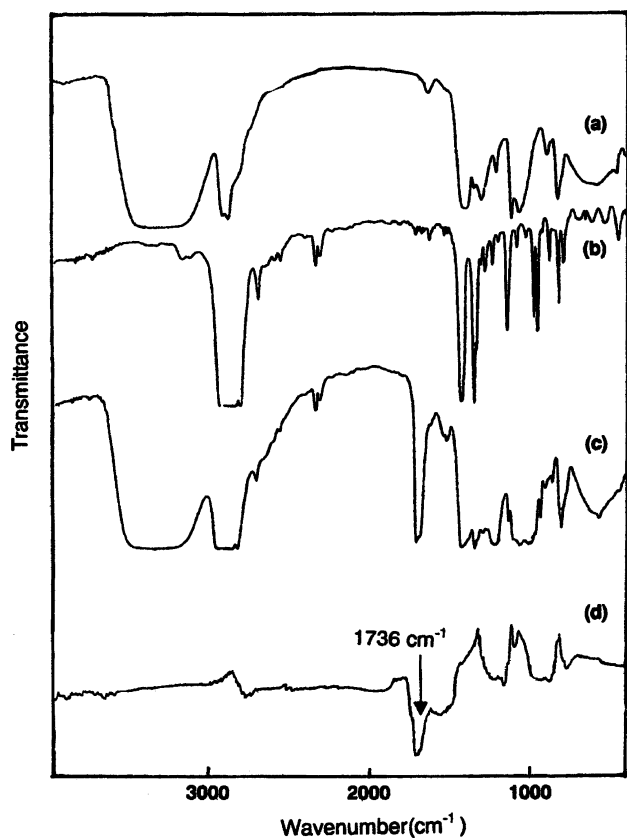
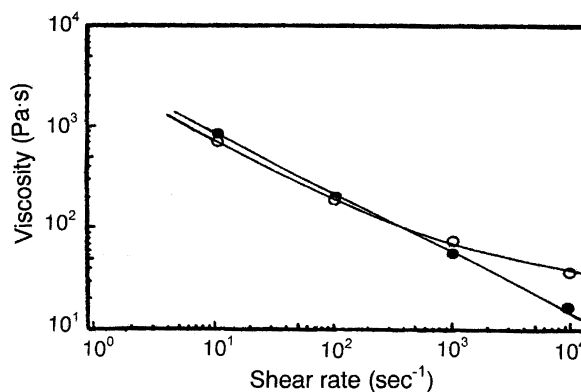
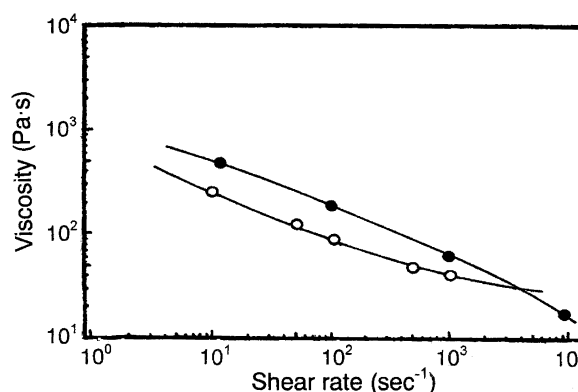


Fig. 1. Fourier transform infrared (FT-IR) spectrum: (a) polyvinyl alcohol; (b) polypropylene; (c) PP/PVA blend; (d) subtraction of the spectra of (a) and (b) from that of (c).



(a)



(b)

Fig. 2. Shear viscosity of PP (●) and PP/PVA (= 50/50 wt%) blend (○) as a function of shear rate: (a) 190 °C; (b) 230 °C.

than that of PP resin, and the viscosity of PP and PVA are depend on the shear rate. Therefore, it can be predicted that the morphology of PP/PVA blend is changed according to a screw speed and extrusion temperature.

In Table 1, the viscosity ratio ( $\eta_{d(PVA)}/\eta_{m(PP)}$ ) of dispersed phase to matrix phase was smaller than unity. In general, when the viscosity ratio ( $\eta_{dispersed\ phase}/\eta_{matrix\ phase}$ ) is smaller or closer to unity, the shear stress may effectively

Table 1  
Melt flow index (M.I.) of polymer according to the measuring temperature and blending ratio

Polymer	M. I. <sup>a</sup> (g/10 min)	
	190 °C	230 °C
PP <sup>b</sup>	1.7	2.5
M.A.-grafted PP	–	34
PP/PVA = 85/15 wt%	–	10.8
PP/PVA = 70/30 wt%	–	7.3
PP/PVA = 50/50 wt%	2.2	4.5
PVA <sup>c</sup>	17.8	45

<sup>a</sup> Load: 2160 g.

<sup>b</sup> PP1: M.I. (g/10 min, 230 °C, 2160 g) = 2.5, density (g/mL) = 0.90.

<sup>c</sup> PVA 205: impregnated with 30 phr of glycerin.

transfer from the matrix phase to the dispersed phase, and the dispersed phase is more easily deformed [7,12,13]. According to measuring temperature, the variation for M.I of PVA was larger than that of PP. Judging from this result, the viscosity ratio ( $\eta_{d(PVA)}/\eta_{m(PP)}$ ) and morphology of PP/PVA blend are largely changed as a function of the extrusion temperature.

### 3.2. Blend morphology and effect of PVA amount

The morphology of a casting PP/PVA (= 70/30 wt%) blend sheet was shown in Fig. 3. 30 phr of glycerin and 0.1 phr of initiator were used with respect to 100 phr of PVA and PP. The casting sheet was not stretched and the thickness was 800–1000  $\mu\text{m}$ . In Fig. 3, the dispersed PVA phase existed as a long fiber form. Fig. 3(a) was the cross

sectional view of PP/PVA blend film (sheet) before stretching. The white specks were PVA fibers and the spot means that a PVA fiber was pulled out during the fracture process of a blend sample in liquid nitrogen. Fig. 3(b) was the perpendicular view to Fig. 3(a). From the figure, the traces of pull-out and hanging PVA fibers also appeared in matrix phase. These long fibers could be formed by the extensional flow and never affected with the take-off speed, because the ratio between extrusion and winding speed was fixed at 1. Therefore, it could be predicted that these long fibers were determined by the low viscosity ratio ( $\eta_{d(PVA)}/\eta_{m(PP)}$ ). In addition, the aspect ratio of PVA fibers was too large to measure compared with the reported results [7,9], as shown in Fig. 3(b). Here, the aspect ratio is defined as the length (major axis of ellipse) of the domain divided by a width (minor axis of ellipse) and characterizes the

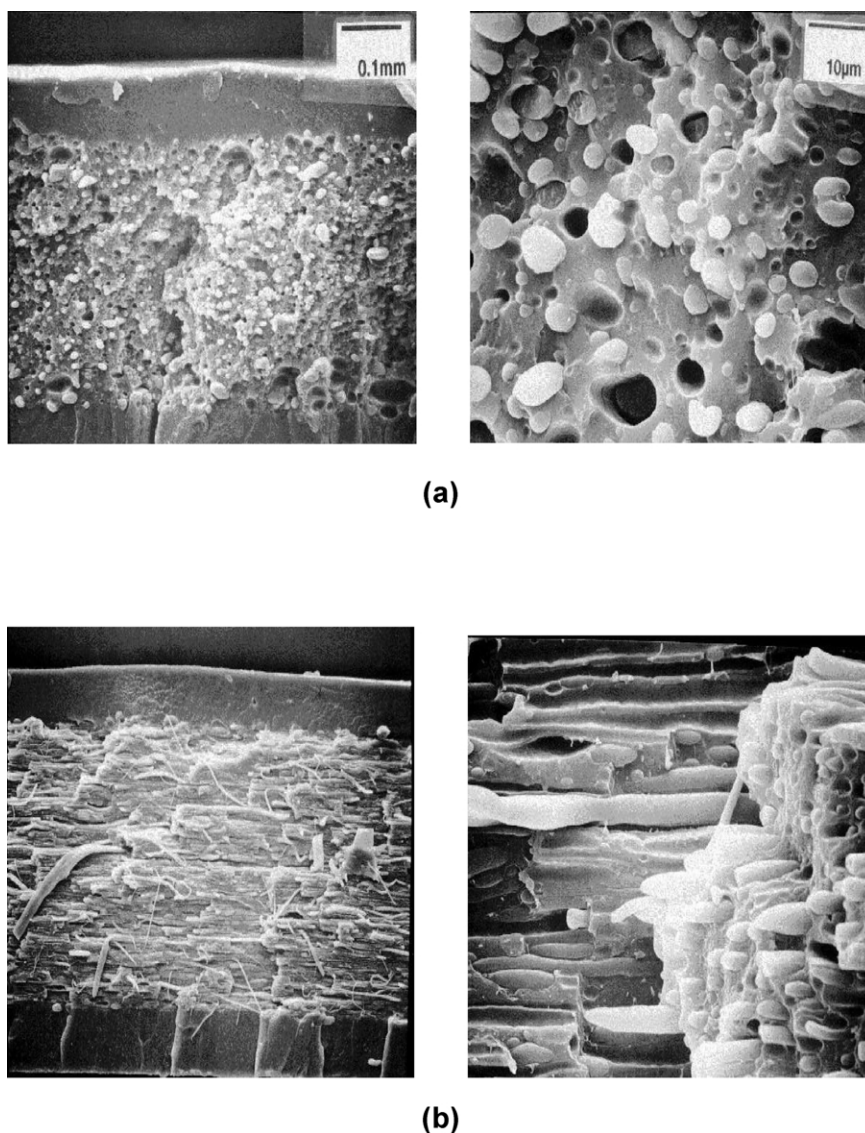
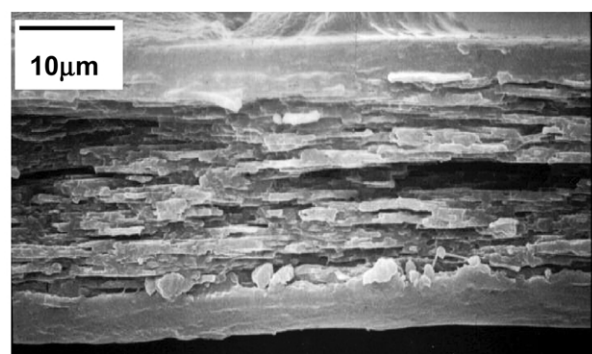


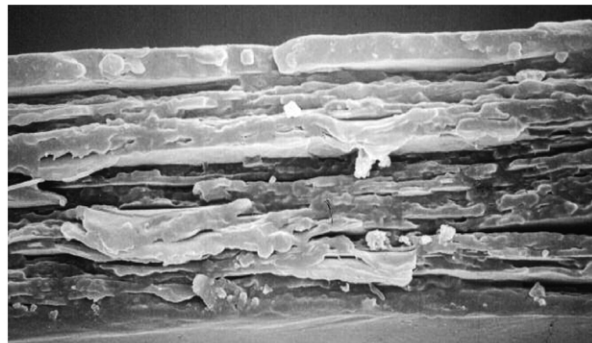
Fig. 3. SEM photomicrographs of the fracture surfaces of extruded PP/PVA (= 70/30 wt%) blend casting sheet: (a) cross-sectional view of a casting sheet; (b) perpendicular view to (a).

extension of deformation [7,10]. This morphology may be advantageous for deforming into platelets when two-dimensional stretching.

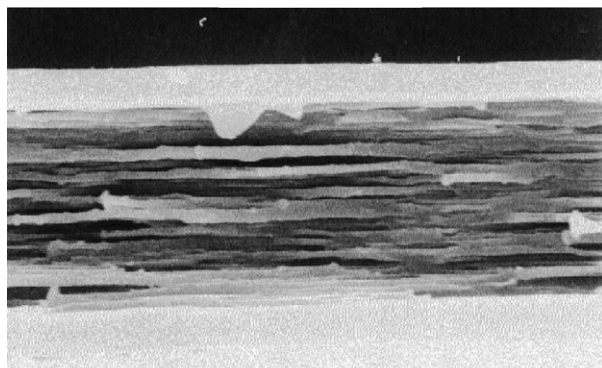
Fig. 4 illustrated the morphology of biaxially oriented PP/PVA blend film with 20–25  $\mu\text{m}$  of thickness. The blend films of Fig. 4(a) and (a') were stretched from a lab. stretcher as a function of the amount of PVA. The PP/PVA (= 70/30 wt%) blend film of Fig. 4(b) was oriented through a BOPP film production line. 30 phr of glycerin and 0.1 phr of initiator were used with respect to 100 phr of PVA and PP. The PVA platelets were developed, as displayed in Fig. 4. In



(a)



(a')



(b)

Fig. 4. SEM photomicrographs of the fracture surfaces of biaxially oriented PP/PVA (TD  $\times$  MD = 8  $\times$  4.5 blend films): (a) PP/PVA = 50/50 wt%, lab. stretching, (a') PP/PVA = 70/30 wt%, lab. stretching; (b) PP/PVA = 70/30 wt%, mass production line stretching.

particular, Fig. 4(b) represented a well-developed laminar morphology, which was similar to multi-layer film or laminated film. In Fig. 4, the black line was a place where a PVA platelet was drew out when the blend sample was fractured in liquid nitrogen. From Figs. 3 and 4, it was confirmed that a PVA domain was deformed from fiber to platelet during biaxially orientation. It could be predicted that the long fibers of cast sheet (Fig. 3) were laterally extended and/or combined each other, and various sizes of platelets were formed and stacked. Table 1 indicated that the viscosity ratio ( $\eta_{d(\text{PVA})}/\eta_{m(\text{PP})}$ ) of PP/PVA blend was largely smaller than unity. Therefore, the deformation of a PVA phase easily occurred by the transferred stress from matrix PP phase during biaxially orientation process.

In Fig. 5, the oxygen permeability and moisture vapor transmission rate (MVTR) of PP/PVA blend film showed as a function of the amount of PVA. However, in case of above 50 wt% of PVA, the blend film was not prepared because of the rupture of blend film during two-dimensional stretching. When the amount of PVA increased, the oxygen permeability was reduced. This result can be predicted that the number of PVA platelet increased with increasing the amount of PVA, and a permeating path of a permeant molecule was elongated. However, the barrier property was not sensitively changed in above 25 wt% of PVA. In addition, Fig. 4(a) and (a') represented the SEM photomicrographs of the fracture surfaces of biaxially oriented PP/PVA blend films having 30 and 50 wt% of PVA. The PVA fibers did not exist, and only PVA platelets existed because of biaxially orientation. When the amount of PVA was 30 wt%, the size of platelets was larger, and the number of platelets was smaller than that of 50 wt% of PVA. It can be judged that the number and size of platelets were varied

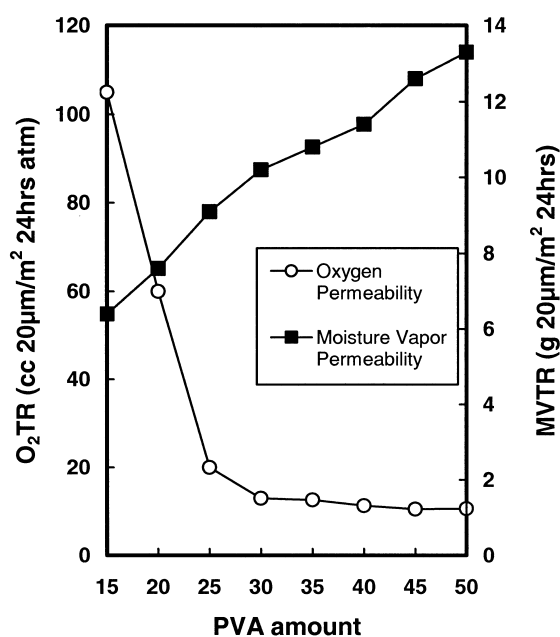


Fig. 5. Oxygen transmission rate and moisture vapor transmission rate of PP/PVA blend films as a function of the amount of PVA.

according to the amount of PVA due to the variation of interfacial tension between PVA and PP. In general, the permeating path of a permeant molecule was determined by the size and number of platelet. Therefore, the oxygen permeability of blend film was changed hardly between 30 and 50 wt% of PVA. MVTR of biaxially oriented PP/PVA film increased with increasing the amount of PVA. In other words, MVTR was not affect with the blend morphology.

The M.I. of PP/PVA blend as a function of blending ratio was illustrated in Table 1. When the amount of PVA increased, the M.I. of blend was reduced. This result can be predicted that the chemical bonding between PP and PVA increased with increasing the amount of PVA. Therefore, the molecular weight of blend became larger because of the grafting reaction between PP and PVA. As a result, the morphology and barrier property as well as M.I. of PP/PVA blend are dependent on the M.I. and blending ratio of PVA and PP.

In Table 2, the oxygen barrier property of the biaxially oriented PP/PVA (= 70/30 wt%) blend film was higher by about 130 times than that of a BOPP film. Compared with the previous reports [7,9,14], it can be judged that the largely improved oxygen barrier property was obtained due to the well-developed platelets.

### 3.3. Effect of plasticizer amount

In case of PVA, the melting point and viscosity were changed according to the type and amount of a plasticizer. Several studies had been performed in order to obtain a good quality film using a plasticizer [17,18].

Table 3 indicated the melting peak temperature ( $T_m$ ) and M.I. of PVA as a function of the amount of glycerin. The M.I. of PVA was in proportion to the amount of a plasticizer. However, the diminished magnitude of  $T_m$  was gradually smaller with increasing the amount of glycerin due to the occurrence of phase separation. Therefore, the efficiency of a plasticizer became lower with respect to PVA. This result is coincided with the oxygen barrier property of PP/PVA (= 70/30 wt%) blend, as shown in Fig. 6.

Fig. 6 showed the oxygen permeability and delamination strength of PP/PVA (= 70/30 wt%) blend film as a function of the amount of glycerin. 0.1 phr of initiator was used with

Table 2  
O<sub>2</sub>TR and MVTR of BOPP and PP/PVA blend film (thickness: 20  $\mu$ m)

Film type	O <sub>2</sub> TR <sup>a</sup>	MVTR <sup>b</sup>
Biaxially oriented PP <sup>c</sup> film	1750	7.3
Biaxially oriented PP <sup>c</sup> /PVA <sup>d</sup> = 70/30 wt% blend film	12.5	9.4

<sup>a</sup> Oxygen transmission rate unit: cc 20  $\mu$ m/m<sup>2</sup> 24 h atm, 25 °C, dry state.

<sup>b</sup> Moisture vapor transmission rate unit: g 20  $\mu$ m/m<sup>2</sup> 24 h atm, 25 °C, 100% RH.

<sup>c</sup> PP1: M.I. (g/10 min, 230 °C, 2160 g) = 2.5, density (g/mL) = 0.90.

<sup>d</sup> PVA 205: impregnated with glycerin 30 phr.

Table 3  
Melting point and melt flow index of PVA as a function of glycerin amount

Glycerin amount (phr)	Melting point (°C)	M.I. <sup>a</sup> (g/10 min)
10	181	17
20	172	32
30	164	45
40	158	>65

PVA 205: dry at 95 °C  $\times$  3 h in a dehumidifying dryer.

<sup>a</sup> Load: 2160 g, temperature: 230 °C.

respect to 100 phr of PP. When the amount of a plasticizer increased, the oxygen permeability was reduced. However, the oxygen permeability increased again at 40 phr of glycerin. This result can be interpreted that the draw extension or deformation of PVA phase diminishes at the decrement of an amount of plasticizer during stretching because of the increment of melting point and viscosity. Therefore, the small platelets and short permeating pathway appear and the barrier property decreases. On the contrary, the surplus glycerin existed in the inner of PVA phase and the degree of crystallization of PVA was downed. Therefore, the diffusion of a permeant molecule was easily performed through the free volume, whereby, the barrier property decreased. In addition, the delamination occurred at the lower force level with increasing the amount of a plasticizer. The delamination happened in the interface between skin and core layers. This phenomenon can be predicted that the plasticizer is continuously diffused from core layer to interface by an applied pressure and temperature during biaxially orientation. When the glycerin absorbed moisture, the possibility of diffusion was raised due to the boiling point ( $T_b$ ) lowering. Although the  $T_b$  of glycerin was 290 °C, it was shifted up to 138 °C at 10% of

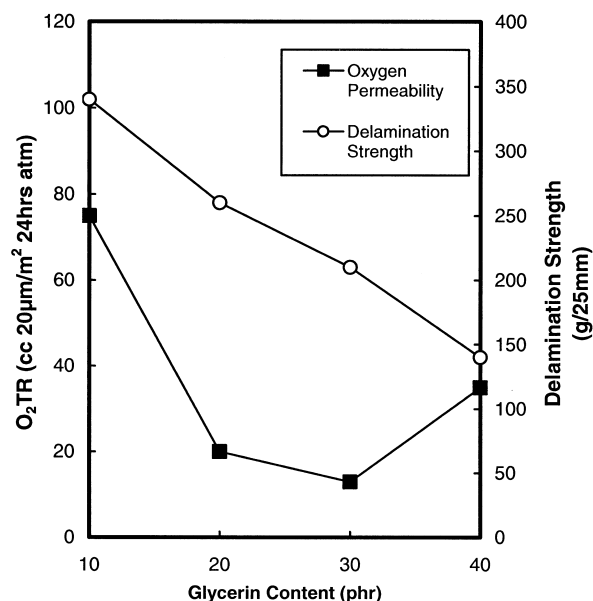


Fig. 6. Oxygen transmission rate and delamination strength of PP/PVA (= 70/30 wt%) blend films as a function of the amount of glycerin.

absorbed moisture. When a biaxially oriented 3-layer PP/PVA blend film was exposed to UV lamp or placed for long time, the delamination strength increased by about 20–30%. This result indicates that the glycerin existing in the interface is continuously evaporated through a skin layer. As a result, it is needed to add the optimum amount of a plasticizer in order to enhance the barrier efficiency and delamination strength of PP/PVA blend.

### 3.4. Effect of initiator amount

In this experiment, M.A. was directly grafted to PP resin and reacted with PVA at the same time using a twin screw extruder. The attack of an initiator generates a free radical as well as degradation of PP resin, and the molecular weight of PP resin rapidly diminished due to  $\beta$ -scission. Although the M.I. of PP resin was 2.5 g/10 min, the M.I. of M.A.-grafted PP was 34 g/10 min at 230 °C. In addition, the M.I. of PVA was 45 g/10 min. However, the M.I. of M.A.-grafted PP/PVA blend became smaller than 34 g/10 min with increasing the amount of PVA, as shown in Table 1. In general, PP has not functional group which can react with PVA. This result means that a free radical is first formed in the PP resin by an initiator and the grafting reaction between M.A. and PP resin occurs, and then grafted M.A. functional group react with the OH group of PVA. In other words, it is possible to react PP, M.A. and PVA simultaneously for the increment of a molecular weight of blend, because PP, M.A. and PVA were fed to a twin-screw extruder at the same time. Therefore, the content of free radical is the critical factor for the reaction between PVA and M.A.-grafted PP as well as PP resin and M.A. monomer.

Fig. 7 illustrated the oxygen permeability and M.I. of PP/PVA (= 70/30 wt%) as a function of the amount of an initiator. 30 phr of glycerin was used with respect to 100 phr of PVA. The curve of oxygen permeability showed 'V' shape as a function of the initiator amount. When the initiator amount was too small, the interfacial tension between PP and PVA phases increased. Therefore, the PVA phase was easily cohered, and the number of PVA platelet and oxygen barrier property of PP/PVA blend decreased. The number of PVA platelet and capacity of compatibilization increased with increasing the amount of an initiator. However, the barrier property of PP/PVA blend was reduced due to the decrement of platelets size. In addition, the M.I. of PP/PVA blend was in proportion to the amount of an initiator. This result indicated that the degradation ( $\beta$ -scission) of PP was more active by an attack of initiator, and the degraded PP played the role of a processing agent. Therefore, the M.I. of blend increased at the increment of amount of an initiator.

As a result, the initiator may play the role of a compatibilizer, and the optimum amount of an initiator should be used in order to obtain the blend film with the excellent barrier property.

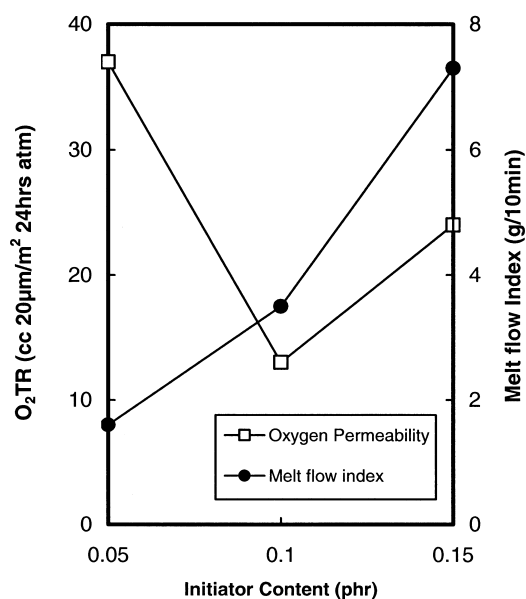


Fig. 7. Oxygen transmission rate and melt flow index of PP/PVA (= 70/30 wt%) blends as a function of the amount of initiator.

### 3.5. Effect of PP viscosity

Fig. 8 displayed the oxygen permeability and delamination strength of PP/PVA (= 70/30 wt%) as a function of the M.I. of PP. 30 phr of glycerin and 0.1 phr of initiator were used with respect to 100 phr of PVA and PP. When the M.I. of PP resin decreased, the oxygen permeability of the PP/PVA blend film was reduced. In other words, the viscosity ratio ( $\eta_{d(PVA)}/\eta_{m(PP)}$ ) of PP/PVA blend diminished, and the thinner and wider platelets were formed. It was known that the dispersed phase was easily deformed by the induced shear stress from the matrix phase with decreasing the viscosity ratio ( $\eta_{dispersed\ phase}/\eta_{matrix\ phase}$ ) [9,11,12,15,16]. In

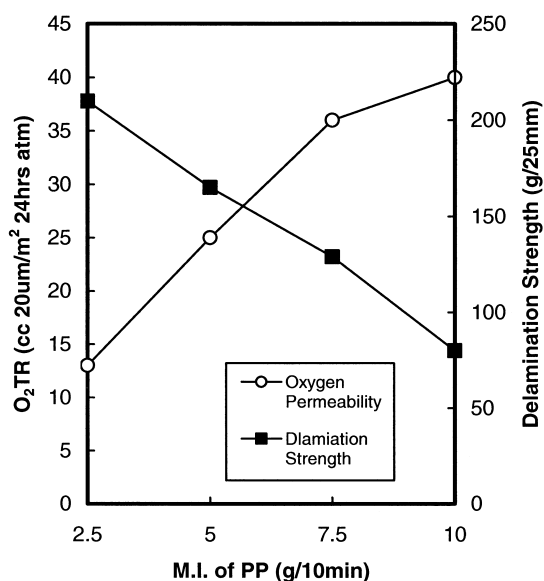


Fig. 8. Oxygen permeability and delamination strength of PP/PVA (= 70/30 wt%) blend films as a function of the melt flow index of PP.

addition, the higher viscosity of matrix PP phase more uniformly transferred the shear stress to a dispersed PVA phase during stretching process. The delamination strength was opposition to the M.I. of PP resin. This result was related to the mechanical property of PP resin. The mechanical property of PP resin was improved with increasing the viscosity, and the delamination did not easily occurred at the lower M.I. of PP.

As a result, the lower M.I. of PP resin is advantages for the oxygen barrier property and delamination of PP/PVA blend film.

### 3.6. Effect of draw ratio

In order to obtain the blend film having an excellent barrier property, the method for forming the laminar structure by means of biaxially orientation has been performed [1,7,9,10,14]. The orientation ratio affects the thickness and area of platelet, and the blend morphology is changed by the draw ratio.

Fig. 9 represented the oxygen and moisture vapor permeability of PP/PVA (= 70/30 wt%) blend film as a function of draw ratio. 30 phr of glycerin and 0.1 phr of initiator were used with respect to 100 phr of PVA and PP. As the draw ratio increased, the oxygen permeability diminished. It could be predicted that the permeating pathway of a permeant molecule was more elongated with increasing the draw ratio, because the size and thickness of PVA platelet were changed according to the draw ratio. The thickness of platelet mainly ranged from 0.15 to 2  $\mu\text{m}$ , and the size (length and width) of platelet was distributed from 4 to 3000  $\mu\text{m}$ . In case of large platelet, the size was larger

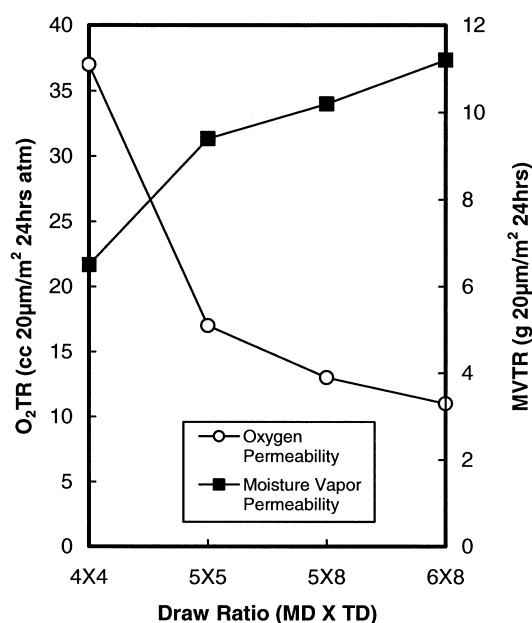


Fig. 9. Oxygen transmission rate and moisture vapor transmission rate of PP/PVA (= 70/30 wt%) blend films as a function of draw ratio.

than approximately 5000  $\mu\text{m}$ . In addition, the minimum draw ratio should be above  $5 \times 5$  in order to obtain an excellent oxygen barrier property.

The moisture vapor permeability was reduced with decreasing the draw ratio. This result means that the moisture vapor permeability is not related to the laminar structure of PP/PVA blend and depends on only the thickness of blend film. The moisture vapor barrier property of PP/PVA blend was fulfilled by the only PP phase due to the high moisture permeability of PVA. Therefore, the moisture vapor permeability of PP/PVA blend film was affected with the thickness of the matrix PP phase, and not related to the PVA platelets.

## 4. Conclusions

The PP/PVA blend film was prepared by means of biaxially orientation process, and the blend morphology and oxygen barrier property has been evaluated as functions of PP/PVA composition and draw ratio.

When the viscosity ratio ( $\eta_{d(\text{PVA})}/\eta_{m(\text{PP})}$ ) of PP/PVA blend was smaller than unity, the PVA phase was transformed from long fibers to the thinner and wider platelets during a biaxially stretching process. The excellent oxygen barrier property of PP/PVA blend film was obtained at the decrement of viscosity ratio, the higher amount of PVA, the optimum amount of plasticizer and initiator, and the increment of draw ratio. However, the oxygen permeability was changed hardly between 30 and 50 wt% of PVA, because the number and size of platelets were varied according to the amount of PVA. The size of PVA platelets was reduced and the number increased at the higher initiator amount, because an initiator played the role of a compatibilizer. Although the laminar structure was developed with increasing the amount of glycerin, the degree of crystallization was reduced due to a surplus glycerin. When the viscosity of PP resin increased, the shear stress induced by the stretching process was more largely and uniformly transferred to PVA phase. The moisture vapor permeability depended on the thickness of the matrix PP phase, and was not related to the laminar morphology of PP/PVA blend due to the high moisture vapor permeability of PVA.

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