Effect of anti-block particles on oxygen transmission rate of SiOx barrier coatings deposited by PECVD on PET films

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A collaboration between:
Industrial PECVD Equipment at Tetra Pak

Typical OTR-Values:
Plain PET 12 μm
120 cm³/m²/day/atm
PET/SiOx (15 nm)
< 3 cm³/m²/day/atm

= Electrical Power + Gas mixture (Disiloxane, Oxygen)
®Hostaphan RNK

Universal film with low haze

®Hostaphan RNK is a highly transparent, biaxially oriented, co-extruded film, made of Polyethylene Terephthalate (PET) and characterized by out-standing physical properties.

Layer structure ®Hostaphan RNK
®Hostaphan RD

Film with one very smooth and glossy surface

®Hostaphan RD is a highly transparent, biaxially oriented, co-extruded film, made of Polyethylene Terephthalate (PET) with different topography of the two surfaces. While the surface structure of one surface is the same as a standard PRT film, the functional surface side displays an extremely regular surface structure with very low roughness.

Layer structure ®Hostaphan RD
PET Film Characterisation
Reflexion Optical Microscopy ($490 \times 330 \ \mu m^2$)

- RNK12
- RD12
- RDO12
Defects on PET Films

Size dispersion of anti-blocking silica and dust particles

Defects of various sizes | Silica Anti-blocking | Dust particles
---|---|---
0 - 15 µm | 0 - 2 µm | 0 - 3.2 µm

Data from tapping AFM: Amplitude
Size Distribution of Anti-blocking particles

![Graph showing size distribution of anti-blocking particles](image-url)
OTR as a Function of Coating Thickness

![Graph showing OTR as a function of coating thickness.](attachment:graph.png)
Effect of Anti-blocking Particles on Oxygen Transmission Rate (OTR) for 40-nm SiOx

<table>
<thead>
<tr>
<th>Film</th>
<th>Particles per mm$^{-2}$</th>
<th>OTR* (cm$^3$/m$^2$/day/atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RNK12</td>
<td>1710</td>
<td>0.58 ± 0.1</td>
</tr>
<tr>
<td>RD12</td>
<td>860</td>
<td>0.39 ± 0.1</td>
</tr>
<tr>
<td>RDO12</td>
<td>290</td>
<td>0.21 ± 0.1</td>
</tr>
</tbody>
</table>

* 23 °C, 50% RH
Fragmentation Tests for SiOx Coatings

9-nm Thick

Crack Density (µm) \(^{-1}\)

Nominal Strain %

COS: 3.8% RNK12
4.8% RDO12

40-nm Thick

Crack Density (µm) \(^{-1}\)

Nominal Strain %

COS: 2.3% RNK12
1.9% RDO12
Durability of SiOx Coated PET Films

The durability of the coating relies on:
- Failure controlled by the coating cohesion strength
- Delamination controlled by adhesion to substrate
- Stress transfer at the coating/substrate interface
Interfacial Shear Strength (IFSS) or Adhesion Strength

The coating’s fragmentation process provides the mean fragment length at saturation, $l_s$, and the coating tensile strength $\sigma_{\text{max}}$ at critical length $l_c = (2/3)l_s$, from which is modeled the interfacial shear strength:

$$\tau = 1.337 \left( \frac{h}{l_s} \right) \sigma_{\text{max}}(l_c)$$
Cohesive Strength

The coating strength is modeled from a two parameters Weibull distribution:

\[ \sigma_{\max} (l_c) = \beta \left( \frac{l_c h}{l_0 h_0} \right)^{-1/\alpha} \cdot \Gamma \{1 + 1/\alpha\} \]

The coating crack onset is derived from the theory of linear elastic fracture mechanics:

\[ \varepsilon_f = \frac{C}{\sqrt{h_c \left( h_s + h_c \right)}} \]
## Mechanical Properties

### 9-nm Thick SiOx

<table>
<thead>
<tr>
<th>Film</th>
<th>COS (%)</th>
<th>$\sigma_{\text{max}}$ (GPa)</th>
<th>$\tau$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RNK12</td>
<td>3.8 ± 0.05</td>
<td>9.5</td>
<td>137</td>
</tr>
<tr>
<td>RDO12</td>
<td>4.8 ± 0.05</td>
<td>7.4</td>
<td>128</td>
</tr>
</tbody>
</table>

### 40-nm Thick SiOx

<table>
<thead>
<tr>
<th>Film</th>
<th>COS (%)</th>
<th>$\sigma_{\text{max}}$ (GPa)</th>
<th>$\tau$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RNK12</td>
<td>2.3 ± 0.05</td>
<td>2.8</td>
<td>94</td>
</tr>
<tr>
<td>RDO12</td>
<td>1.9 ± 0.05</td>
<td>4.7</td>
<td>196</td>
</tr>
</tbody>
</table>
Conclusion

1. Oxygen transmission rate (OTR) and crack onset (COS) of PECVD SiOx depend on the density of anti-block particles at bi-oriented PET film surfaces.

2. OTR is proportional to the surface density of anti-blocking particles.

3. COS increased up by 20% for substrate with the lower anti-blocks density.

4. Micro-mechanical modeling of fragmentation process of SiOx coatings provided evaluation of their cohesive and adhesion strengths on PET.

5. Cohesion force of 9-nm thick SiOx is twice higher than 40-nm coatings.

6. Interfacial shear strength is as high as the bulk shear stress reflecting the presence of covalent bonds at the interface.
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