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Slip and Roughness at a ETFE/PS Interface during Coextrusion

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Abstract: We investigated the relationship between interfacial slip and interfacial roughness during the coextrusion of ethylene-tetrafluoroethylene (ETFE) and Polystyrene (PS) in a capillary rheometer. The interfacial slip velocity was determined using the modified Mooney method and the interfacial roughness was evaluated using a laser microscope. The interfacial slip velocity exhibited a power-law dependence on the interfacial stress. At low shear stresses, the power-law exponent was approximately 6. Beyond a critical interfacial stress, $\sigma_{c\text{-}i}$, the power-law exponent decreased to value of approximately 2. At around the same critical interfacial stress, $\sigma_{c\text{-}i}$, interfacial roughness also begins to increase with the interfacial shear stress. This suggests that the interfacial slip and interfacial roughness occur together. However, by comparing coextrusion flow in dies with and without a contraction, it appears that both interfacial slip and contraction flow are needed for the observation of interfacial roughness.

Keywords: “Coextrusion” , “Interfacial slip” , “Interfacial roughness”, “ETFE”, “PS”.

1. Introduction

Coextrusion processing offers great advantages including the ability to readily adjust the number of layers, control the thickness of the layers etc. It can often be a lower cost option than other techniques for producing multilayers, such as lamination, coating etc.. However, multilayers produced by coextrusion often exhibit roughness at the polymer-polymer interface. This roughness significantly worsens the appearance of the multilayer and also has a deleterious effect on physical properties. Past work suggests that the occurrence of interfacial roughness is strongly correlated to the occurrence of interfacial slip.

Recently, Komuro et al.[1] proposed for a modification of the Mooney method, typically used to investigate wall slip, to investigate slip at a polymer-polymer interface. They found that interfacial slip velocity varies with the interfacial stress as a power-law, and that the power-law exponent changed at a critical stress σ_i.[2]

In this study, we focused on the interfacial slip and the interfacial roughness during the coextrusion of Ethylene-tetrafluoroethylene copolymer (ETFE) and PS using a capillary rheometer.

2. Materials and Method

2.1 Materials

We used ETFE (Asahi Glass, Tm=190℃) and PS (PS Japan, Mw=312,000). An ETFE rod of length 150 mm length and diameter 9.2 mm was prepared by extrusion. In order to coat a thin layer of PS on ETFE, the ETFE rod was first dipped in a 20wt% PS/tetrahydrofuran (THF) solution. The coated rod was then dried in a vacuum oven at 80℃ for 1 week.

2.2 Measuring the polymer-polymer interfacial slip velocity

The core-shell coated rod sample was coextruded in a capillary rheometer (Capilograph, Toyoseiki). Capillary dies of three different diameters D=1.0, 1.5 and 2.0 were used. The die length to diameter ratio L/D=20 and the entry angle was fixed at 60°. The volume flow rate at different interfacial stress values was measured. The modified Mooney method of Komuro et al.[1, 2] was used to determine the slip velocity as a function of the interfacial shear stress.

2.3 Interfacial stress calculation by numerical simulation (2 dimensions, generalised Newtonian fluid)

Interfacial slip velocity depends on the interfacial stress. In order to determine the coextrusion conditions corresponding to the observation of interfacial roughness, we performed numerical simulations of a 2 dimensional generalised Newtonian fluid. The chosen coextrusion conditions corresponded to a range of interfacial shear stresses that included the critical interfacial stress, $\sigma_{c\text{-}i}$.

2.4 Coextrusion of ETFE/PS multilayers

The coextrusion conditions determined using numerical simulation was used to fabricate the ETFE/PS multilayers. The existence or non-existence of interfacial roughness at different values of the interfacial stress was determined.

3. Results and Discussion

3.1 Slip velocity and interfacial roughness by capillary coextrusion.

The interfacial shear stress dependence of the slip velocity is shown in Fig. 1. As can be seen from Fig.1, the dependence of the interfacial slip velocity on the interfacial shear stress is a power-law.

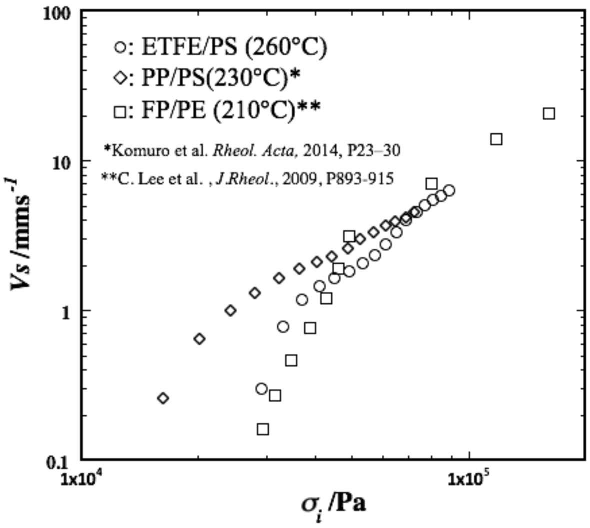

Fig.1 Slip velocity at the ETFE/PS interface at T=260℃. For comparison, literature data from two other polymer pairs are also shown.

Consistent with previous work on similar polymers[2,3], the power-law exponent was approximately 6 at low shear stresses and decreased to approximately 2 at a critical interfacial shear stress, $\sigma_{c\text{-}i} \approx 4.0\times10^4$ Pa.

In order to investigate the interfacial roughness, the PS layer was first dissolved using THF. The interface was dried and then scanned using a laser microscope.Fig.2 presents the relationship between interfacial roughness and the interfacial shear stress. The interfacial roughness is parametrised by the standard deviation of the positive and negative deviations normal to the interface, R_q. As can be seen from Fig. 2, R_q is essentially constant at low stresses and begins to increase beyond a certain critical stress. Interestingly, this critical stress is essentially identical to the critical stress of the slip velocity, $\sigma_{c\text{-}i}$.

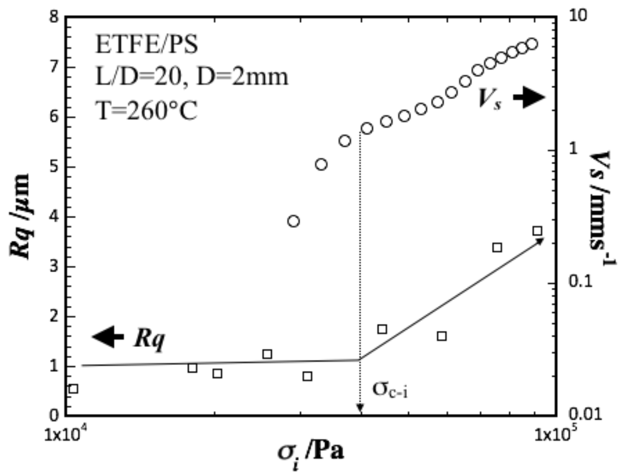

Fig.2 Interfacial slip velocity (V_s) and interfacial roughness at the ETFE/PS interface(T=260℃).

3.2 Calculating of interfacial stress by CAE

The die geometries used are indicated in Fig.3. Using these die geometries the calculated flow rates, pressure drop and the corresponding interfacial stresses are indicated in Table 1(we showed only using geometry (a) calculated in this abstract, and Test2,3,4 on Table 1 indicated over the critical interfacial stress $\sigma_{c\text{-}i}$).

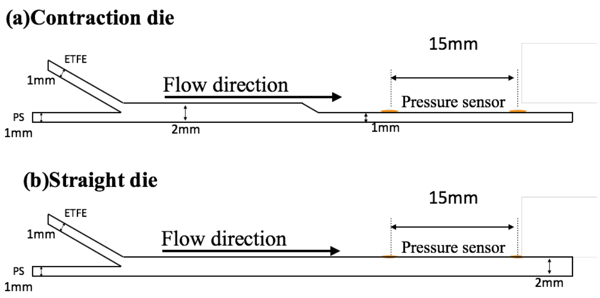

Fig.3 Geometries of the die used in this study.

Table.1 Calculated flow rates, pressure drops and the corresponding interfacial stress for the coextrusion of ETFE/PS through the die indicated in Fig. 3 (a).

Test	$Q_{ETFE}:Q_{PS}$	ΔP(MPa)	$\sigma_i(\times 10^4 Pa)$
1	1:6	1.49	3.0
2	1:9	1.74	4.0
3	1:12	2.00	4.7
4	1:14.7	2.20	5.3

3.3 Interfacial roughness during the coextrusion of ETFE/PS.

ETFE and PS were coextruded at the conditions determined by the numerical simulation. Experimental data in coextrusion using die geometry (a) showed Table 2. At the conditions that corresponded to an interfacial stress that was greater than the critical value, the presence of interfacial roughness was visually confirmed. Visual confirmation is easier using the corresponding movies, rather than the presented photographs.

Table.1 Experimented data of the flow rates, pressure drops and the corresponding interfacial stress for the coextrusion of ETFE/PS through the die indicated in Fig. 3 (a).

Test	$Q_{ETFE}:Q_{PS}$	ΔP(MPa)	$\sigma_i(\times 10^4 Pa)$
1	1:6	1.71	3.0 ~ 4.0
2	1:9	1.9	4.0 ~ 4.7
3	1:14.7	2	4.7 ~ 5.3

4. Conclusions

A brief summary of findings is as follows:

1. The dependence of the interfacial slip velocity on the interfacial shear stress during the coextrusion of ETFE/PS is a power-law. The power-law exponent of approximately 6 at low shear stresses decreases to approximately 2 at a critical interfacial stress, $\sigma_{c\text{-}i} \approx 4.0 \times 10^4$ Pa

2. Interfacial roughness was essentially negligible at low shear stresses and begins to increase at the same critical stress corresponding to the interfacial slip velocity, $\sigma_{c\text{-}i}$. Therefore, we suggest that interfacial slip and interfacial roughness are related.

3. We confirmed that interfacial roughness was observed when the interfacial stress was greater than the critical interfacial stress $\sigma_{c\text{-}i}$, provided the die included a contraction.

References

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