# A Comparison of Melt Fracture Initiation Conditions in Capillaries and Slits

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#### **Synopsis**

An experimental study was carried out with the objective of comparing the critical conditions for the onset of gross extrudate distortion, usually called melt fracture, in capillaries and slits. Narrow and broad molecular weight distribution polystyrenes as well as low- and high-density polyethylenes were used. The onset of melt fracture was observed to take place at higher shear stresses in slits than in capillaries. It is argued that the flow-average value of the recoverable shear strain should be used as the criterion for the initiation of melt fracture.

## INTRODUCTION

Whenever a polymer melt is extruded at high shear rates and stresses, the extrudate becomes distorted. This phenomenon is referred to as melt fracture, or melt flow instability. There are numerous papers in the literature devoted to the study of mechanisms responsible for this phenomenon and to the determination of dependence of critical quantities to such variables as die entry geometry, temperature, and molecular weight. There are also many reports of flow visualization studies aimed at determining the initiation site for instability with both solutions and melts. Despite the large amount of experimental and theoretical work, we still do not have a clear understanding of this phenomenon. There is considerable disagreement in the literature, however, most authors agree that melt fracture sets on at a constant value of wall shear stress which is of the order of 10<sup>6</sup> dynes/cm<sup>2</sup> for polymer melts. The corresponding critical recoverable shear strain defined as half the ratio (first normal stress difference/shear stress) has been found to vary between about 1 and 60 units.<sup>1,2</sup> This wide range of values is partly due to the variety of experimental methods used in the determination of recoverable shear and to molecular weight distribution effects. There are two excellent papers by Tordella<sup>3</sup> and White<sup>4</sup> that present a comprehensive review of most of the important experimental work through 1965 and 1972, respectively. Subsequent publications, both experimental and theoretical, are discussed in a comprehensive and critical review of instabilities in polymer processing by Petrie and Denn.<sup>5</sup>

Virtually all experimental work has been carried out with capillary dies of circular cross section. den Otter,<sup>6</sup> however, reports that for silicones, melt fracture occurs at the same shear rate and shear stress in capillaries and slits, within experimental accuracy. Wales<sup>7</sup> reports that for HDPE, the onset of irregular flow takes place at about the same wall shear stress for both capillaries

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and slits, while PS and LDPE fail to show any irregularity in slit flow at wall shear stresses where melt fracture occurred in capillary flow. Petrie and Denn<sup>5</sup> cite three unpublished studies regarding melt fracture behavior in slits. The results are rather inconclusive, and it is not clear whether slit flow is definitely more stable than capillary flow.

Rothenberger, McCoy, and Denn<sup>8</sup> carried out a linear stability analysis of circular and plane Poisseuille flow of a Maxwell fluid in the limit of zero Reynolds number. For the circular cross-sectional geometry, they obtained a critical recoverable shear of 2.6, which agrees well with experimental results for monodisperse polystyrene.<sup>2,9</sup> For the two-dimensional geometry, they obtained a critical recoverable shear of 0.7. More accurate calculations, however, have shown that the critical recoverable shear for two-dimensional Poisseuille flow is too low. $^{5}$ 

<b>Characteristics of Polymers Investigated</b>							
Туре	Trade name	Supplier	$\overline{M}_w^{\mathrm{a}}$	$\overline{\overline{M}_{w}}/\overline{M}_{n}{}^{a}\overline{M}_{z}\overline{M}_{z+1}/\overline{M}_{w}{}^{z}b$			
PS	Styron 456	Dow Chemical					
PS	Lustrex HT 42-1	Monsanto					
PS	Lustrex HF-55	Monsanto	320,000	3.10			
PS	Standard	Pressure Chemical	200,000	<1.06	1.25		
PS	Standard	Pressure Chemical	110,000	<1.06	1.25		
LDPE	DFDY-3701	Union Carbide	51,300	3.72			
LDPE	DFDQ-0118	Union Carbide	53,500	3.00			
LDPE	UFG-1	Union Carbide	70,900	3.63			
LDPE	DFDY-6005	Union Carbide	82,800	3.96			
LDPE	DYNH-5	Union Carbide	88,800	6.25			
LDPE	Lupolen 2000H	BASF					
HDPE	HD-211	Union Carbide	100,500	5.07			
HDPE	DMDJ 4309	Union Carbide	150,000	7.42			

TABLE I

<sup>a</sup>Provided by supplier.

<sup>b</sup>Assumed.

It is the purpose of this paper to report and compare some new experimental results on melt fracture in capillaries and slits. The critical shear stress is measured directly, and the critical recoverable shear is calculated from the critical shear stress values and semiempirical expressions for the shear compliance.

## **EXPERIMENTAL**

## **Materials**

The materials used were polystyrenes (PS), low-density polyethylenes (LDPE), and high-density polyethylenes (HDPE). The polystyrenes included both commercial samples and narrow molecular weight distribution standards. All polyethylenes were of commercial grade. The commercial samples were supplied by the manufacturers in the form of pellets, and the standard polystyrenes were in the form of flakes. The experiments for PS were performed at 160°C, for LDPE at 150°C, and for HDPE at 135°C. The characteristics of all polymer samples used are summarized in Table I. The experimental data for Lupolen 2000 H (LDPE) were supplied by G. Ehrmann.<sup>10</sup>

## **Experimental Procedure**

All experiments were carried out in an Instron Model 3211 capillary rheometer (constant shear rate instrument). The Model 3211 is a totally self-contained unit which includes an extrusion barrel assembly, a push button-operated drive system, a temperature control and distribution system, and an electronic force measuring and readout system. The extrusion barrel assembly consists of a cylindrical reservoir made of hardened steel, 0.375 in. in diameter, enclosed in an aluminum heating jacket. The die is inserted into the bottom of the reservoir and is held by a simple clamping nut.

Dimensions of Capillaries Used (Flat Entry) <sup>a</sup>							
Capillary no.	Diameter D, in.	Length $L$ , in.	L/R				
1	0.064	2.990	93.4				
2	0.063	1.546	49.0				
3	0.068	1.248	39.6				
4	0.063	1.001	31.8				

 TABLE II

 Dimensions of Capillaries Used (Flat Entry)

<sup>a</sup>G. Ehrmann's capillaries (flat entry)  $L/R = 20, 40, 100, D = 1 \text{ mm.}^{10}$ 

Both dies of circular cross section (capillaries) and dies of rectangular cross section (slits) were used in this study. A capillary was formed by drilling a circular hole of a certain diameter through a cylinder of brass nearly 0.375 in. in diameter. A slit was made in a little more complicated manner. First, a hole of diameter approximately 0.3 in. was drilled through a cylinder of brass nearly 0.375 in. in diameter. Another cylinder of brass was machined to a diameter slightly larger than that of the hole. This cylinder was then split into two slightly unequal parts. A channel with the desired rectangular cross section was milled into the surface of the larger part, the other was left flat. The two parts were put back together and pressed into the hole. All capillary and slit dies used had a flat entry (180 degrees). Their dimensions are given in Tables II and III.

In the experiments, two quantities were directly measured, i.e., the plunger speed and the force on the plunger. For the polystyrene samples, except for HT 42-1 and Styron 456, data were obtained from three capillaries of different L/R ratios and three slits of different L/h ratios. The true shear stress at the wall was obtained by performing the usual entrance correction (Bagley plot). For HT 42-1, Sytron 456, and all polyethylene samples, the data were obtained from a single capillary (L/R = 93.4) and a single slit (L/h = 95.3). In subsequent figures and tables, the shear stress determined from a single die (either capillary or slit) is considered as the true shear stress although no entrance correction was performed. It was estimated that apparent shear stresses determined with the dies used were a bit higher than true stresses, but no more than 5% or 10%. Consequently, for comparison purposes (slit vs. capillary), such errors are of little significance. For all polymers tested, the true shear rate at the wall was determined by using the Rabinowitsch equation.

Slit no.	Width $w$ , in.	Thickness h, in.	Length $L$ , in.	w/h	L/h
1	0.24	0.015	1.430	16	95.3
2	0.24	0.015	0.743	16	49.5
3	0.24	0.015	0.602	16	40.1
4	0.24	0.015	0.480	16	32.0

TABLE III Dimensions of Slits Used (Flat Entry)<sup>a</sup>

<sup>a</sup>G. Ehrmann's slit die (rounded, trumpet-like entry) w = 0.52 mm, w/h = 15.6, L/h = 70.<sup>10</sup>

The flow curves ( $\tau_w$  vs.  $\dot{\gamma}_w$ ) obtained with slit dies were virtually identical to those obtained with capillary dies. There was a slight tendency for the slit data to shift to either side of the capillary data in some cases, but the agreement was generally good. Therefore, within experimental error, we can say that capillaries and slits yield a single flow curve for the materials investigated, in agreement with most previous works.<sup>6,11,12</sup> Some typical flow curves are shown in Figures 1–3.



Fig. 1. Logarithmic plot of shear stress vs. shear rate for polystyrene HF-55: ( $\bullet$ ) capillary; ( $\Delta$ ) slit.

### RESULTS

## **Critical Shear Stress**

The onset of melt fracture was determined visually by observing the appearance of the extrudates. When capillaries were used, the extrudates were straight and smooth at low shear rates. When the shear rate was increased, a gross distortion set on at a certain value. This gross distortion was in some cases preceded by surface roughness. The form of distortion was different from polymer to polymer. The polystyrenes exhibited a characteristic waviness and a tendency to form spirals. The low-density polyethylene extrudates were distorted by the appearance of knobby kinks at more or less regular intervals. The high-density polyethylenes exhibited a pronounced waviness.



Fig. 2. Logarithmic plot of shear stress vs. shear rate for low-density polyethylene DFDY-0701: ( $\bullet$ ) capillary; ( $\Delta$ ) slit.

Extrudate distortion in slit extrusion was observed at relatively low flow rates for virtually all samples tested. In slits, the restricting influence of the two edges produces a higher flow rate in the midsection of the die. The material in the middle which is flowing at a higher shear rate than that near the edges will either swell excessively or buckle upon emergence from the slit. Buckling results in a large-amplitude regularly rippled surface like that observed by Tordella.<sup>13</sup> This kind of midsection buckling gradually diminishes when the extrudate is stretched by its own weight, and can be eliminated by attaching a small weight at the leading end of the extrudate. Attaching the same small weight at the leading end of fractured extrudates emerging from capillaries or slits neither eliminated nor reduced the degree of the distortion. Consequently, the disappearance of midsection buckling under small stretching of slit extrudates confirms that this type of distortion is not what is normally referred to as melt fracture.

With PS and HDPE, when the shear rate was gradually increased up to a certain critical value, a distortion transverse to the direction of flow was observed in the form of "ragged edges" and an overall deterioration of extrudate appearance. This kind of distortion persists even under a stretching force, and is taken to be melt fracture in slits by the authors.

In the extrusion of LDPE through slits, another type of flow defect was observed in the form of a "ridge-like" structure that runs perpendicular to the flow direction of the extrudate surface. This kind of defect appears at shear rates below the onset of "ragged edges" (our definition of melt fracture) and persists beyond it. This "ridge-like" structure is possibly the waves and ripples as observed by Tordella<sup>13</sup> and Schott and Kaghan.<sup>14</sup> We feel that this type of defect is more like sharkskin, a surface roughness, than melt fracture, a gross distortion.



Fig. 3. Logarithmic plot of shear stress vs. shear rate for high-density polyethylene HD-211:  $(\bullet)$  capillary;  $(\Delta)$  slit.

From the foregoing discussion, it is apparent that the determination of the onset of melt fracture is to a certain extent subjective. By checking the appearance of the extrudates with several experienced research workers, it was established that the critical shear stress values given in this paper are accurate within about  $\pm 5\%$  for capillary dies. In the extrusion of polymer melts through slits, however, the type and the severity of distortion can easily be confused. Persistence of the midsection buckling and the gradual appearance of surface roughness (in some cases) complicate the determination of the onset of gross melt fracture. The experimental results for slits given in Tables IV, V, and VI represent the conditions for the onset of the "ragged edges" distortion which in our opinion corresponds to what is normally referred to as melt fracture in capillaries. For the LDPE samples, the shear stress at the onset of "ridge-like" surface roughness is also given (Table V). Critical shear stress and critical shear rate values for capillaries and slits are shown side by side for direct comparison. It can be seen that for all samples tested,  $(\tau_{wc})_{slit}$  is higher than  $(\tau_{wc})_{cap}$ , the ratio  $(\tau_{wc})_{slit}/(\tau_{wc})_{cap}$  ranging from 1.15 to 1.62. The smallest value is for Ehrmann's data<sup>10</sup> and may be due to the difficulty in determining the onset of gross distortion and to the fact that the slit had a trumpet-like entry.

styrene at 160°C	Slits	)-6, $\dot{\gamma}wc$ , $\sigma_{wc}(\overline{M_z}\overline{M_w^2}^{z+1})^{-1a} \sigma_{wc}(\overline{M_z}\overline{M_{z^{+1}}})^{-1b} (\sigma_{wc})_{slit}^{a/} (\sigma_{wc})_{slit}^{a/}$ $\overline{M_w^2}$ sec <sup>-1</sup> $(\sigma_{wc})_{cap} (\sigma_{wc})_{cap}$	$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			ABLE V w-Density Polyethylene at 150°C	Slit	$ \begin{array}{llllllllllllllllllllllllllllllllllll$		1.47 743 0.8 1.48	1.87 $927$ $<1.0$ $1.60$	0.981 154 0.6 1.45	0.888    40.1    <0.4    1.57	0.772 116 < $0.367$ 1.57	$0.814$ 150 $\sim 0.706$ 1.15	
		$(\sigma_{wc})$	adada			ļ		-' at urface	ess, m <sup>2</sup>					7	9	
		$\sigma_{wc}(\overline{\frac{M_z}{M_{w^2}}})^{-}$	4.04 3.94 6.14 					$ au_{wc}  imes 10^{-}$ onset of su	roughne dynes/c	0.8	<1.0	0.6	< 0.4	< 0.36	~ 0.70	
	Slits	$J_{wc}(\overline{\frac{M}{M}}_{z}\overline{M}_{w^{2}})^{-1}a$	2.67 1.88 1.56		e at 150°C Stit	Slit	Slit	$\dot{\gamma}_{wc}$ , sec <sup>-1</sup>	743	927	154	40.1	116	150		
ie at 160		Ýwc, ( sec-1	103 43.5 401 102 38.5			/ethylene			$10^{-6}$ ; s/cm <sup>2</sup>	7	7	81	88	72	14	
TABLE IV Critical Conditions for Polystyren		$ au_{wc}  imes 10^{-6}$ , dynes/cm <sup>2</sup>	3.26 2.01 1.54 1.67 2.22		r V nsity Poly			$\tau_{wc} \times dyne$	1.4	1.8	0.9	0.8	0.7	0.8		
	Capillaries	$\sigma_{wc}(\frac{\overline{M}_{z}\overline{M}_{z+1}}{\overline{M}_{w^{2}}})^{-1}b$	2.40 2.53 3.55			TABL1 itions for Low-De		llary	$\dot{\gamma}_{wc}, \sec^{-1}$	298	297	57.7	9.73	35.8	104	(170°C). <sup>10</sup>
		$\sigma_{wc}(\frac{\overline{M_zM_z+1}}{\overline{M_w^2}})^{-1}a$	1.64 1.35 1.03 			Critical Cond		Capi	$ au_{wc}  imes 10^{-6}$ , dynes/cm <sup>2</sup>	0.997	1.17	0.678	0.565	0.491	0.706	l by G. Ehrmann
		6, Ϋ <i>wc</i> , n² sec <sup>-1</sup>	44.5 13.9 80.3 41.7 14.9						10-4	13	35	60	28	88	I	supplied
		$\tau_{wc} \times 10^{-}$ dynes/cn	2.01 1.44 1.02 1.17 1.17						$\overline{M}_{w} \times$	5.	5.0	7.1	õ	80	1	ner were
		$M_w  imes 10^{-5}$	1.1 3.2 1.1	q. (4). q. (7).					name	-0701	-0118		-6005	1-5	en 0H <sup>a</sup>	r this polyn
		Trade name	Narrow MWD Narrow MWD HF-55 HT 42-1 Styron 456	<sup>a</sup> From e <sup>b</sup> From e					Trade	DFDY	DFDQ	UFG-1	DFDY	DYNH	Lupol 2001	<sup>a</sup> Data fo

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#### **Critical Recoverable Shear Strain**

Hooke's law in shear can be written

$$\sigma_w = \tau_w J \tag{1}$$

where  $\tau_w$  is the shear stress at the wall, J is the "true" shear compliance, and  $\sigma_w$  is the recoverable shear strain at the wall.

The steady-state Rouse shear compliance of a collection of flexible chains is given by  $Ferry^{15}$  as

$$J_{\rm R} = \frac{2}{5} \left( \frac{\overline{M} r_w}{\rho R T} \right) \left( \frac{\overline{M}_z \overline{M}_{z+1}}{\overline{M}_w^2} \right) \tag{2}$$

where  $\rho$  is the density, R is the ideal gas constant, T is the absolute temperature, and  $\overline{M}_w$ ,  $\overline{M}_z$ ,  $\overline{M}_{z+1}$  are the weight, z, and z + 1 average molecular weights.

A correlation between the "true" steady-state shear compliance at zero shear,  $J_0$ , and the Rouse compliance was developed by Graessley,<sup>16,17</sup> which is

$$\frac{J_0}{J_{\rm R}} = \frac{A}{1 + B\rho E_0} \tag{3}$$

where  $E_0 = M_w/16,500$  is the entanglement density at zero shear, A = 2.2 and B = 0.347 for polystyrene.

By substituting J with  $J_0$  in eq. (1) and assuming this to be valid up to  $\tau_{wc}$ , we have

$$\sigma_{wc} = \tau_{wc} \frac{J_{\rm R}A}{1 + B\rho E_0} \tag{4}$$

Provided that both capillary and slit measurements are carried out at the same temperature, the ratio of the critical recoverable shear at the wall of a slit die to the corresponding value in capillary extrusion is simply

$$(\sigma_{wc})_{\rm slit} / (\sigma_{wc})_{\rm cap} = (\tau_{wc})_{\rm slit} / (\tau_{wc})_{\rm cap}$$
(5)

We have assumed, thus far, that the shear compliance is independent of shear rate, and we have used Graessley's zero shear value for the calculation of recoverable shear strain at the onset of melt fracture. We may allow for changes in shear compliance by replacing the entanglement density at zero shear,  $E_0$ , by the entanglement density at finite shear rates,  $^{16,18}E(\dot{\gamma})$ , so that

$$\frac{J(\dot{\gamma})}{J_{\rm R}} = \frac{A}{1 + B\rho E(\dot{\gamma})} \tag{6}$$

 $E(\dot{\gamma})$  can be calculated from Graessley's entanglement theory using a monodisperse polymer approximation as suggested by Vlachopoulos et al.<sup>18</sup> By substi-

TABLE VI Critical Conditions for High-Density Polyethylene at  $135^{\circ}C$ 

		Capil	llary	S			
Trade name	$M_w$	$ au_{wc}, \\ dynes/cm^2$	$\dot{\gamma}_{wc}, \sec^{-1}$	$\tau_{wc}$ , dynes/cm <sup>2</sup>	$\dot{\gamma}_{wc}$ , sec <sup>-1</sup>	$(\sigma_{wc})_{ m slit}/ (\sigma_{wc})_{ m cap}$	
HP-211 DMDJ 4309	100,500 150,000	$\begin{array}{c} 0.851 \\ 0.743 \end{array}$	$\begin{array}{c} 113\\27.9\end{array}$	$\begin{array}{c} 1.16\\ 1.05\end{array}$	$\begin{array}{c} 177\\ 44.4\end{array}$	1.36 1.41	

tuting J with  $J(\dot{\gamma})$  in eq. (1), we have

$$\sigma_{wc} = \tau_{wc} \frac{AJ_{\rm R}}{1 + B\rho E(\dot{\gamma})} \tag{7}$$

Then, the ratio of the critical values of the recoverable shear strain is

$$\frac{(\sigma_{wc})_{\text{slit}}}{(\sigma_{wc})_{\text{cap}}} = \frac{(\tau_{wc})_{\text{slit}}}{(\tau_{wc})_{\text{cap}}} \frac{1 + B\rho E(\dot{\gamma}_c)_{\text{cap}}}{1 + B\rho E(\dot{\gamma}_c)_{\text{slit}}}$$
(8)

For polystyrene, the critical values of recoverable shear using eqs. (4) and (7) are tabulated in Table IV. It can be seen that the method of calculation does affect the absolute values of the recoverable shear, but the ratio  $(\sigma_{wc})_{slit}/(\sigma_{wc})_{cap}$  is only slightly affected. For LDPE and HDPE, the calculations are based on eq. (5) only because of lack of correlations analogous to eq. (6) for these polymers.

It can be seen from Tables IV to VI that  $(\sigma_{wc})_{slit} > (\sigma_{wc})_{cap}$  for all the samples tested, whether a shear rate-independent  $J_0$  or a shear rate-dependent  $J(\dot{\gamma})$  is used in the calculations. The determination of  $\sigma_{wc}$  in terms of Graessley's correlation is essentially an extrapolation of rheogoniometer data for  $\dot{\gamma} \rightarrow 0$  to high rates of shear. Whether such an extrapolation is valid by using either  $J_0$  or  $J(\dot{\gamma})$ is still subject to question and needs further experimental verification. There is also some evidence<sup>19</sup> that the shear compliance J might actually decrease with  $\dot{\gamma}$  for polydisperse PS rather than increase as indicated by eq. (6). Consequently, the absolute values of the recoverable shear given in this paper might not be very accurate. However, the ratios  $(\sigma_{wc})_{slit}/(\sigma_{wc})_{cap}$  are probably correct within the experimental error for the determination of the corresponding critical shear stresses.

In the foregoing discussion, the critical recoverable shear strain is calculated at the wall of the capillary or slit die. There is no assumption made as to the mechanism involved or the initiation site. This recoverable shear strain hypothesis has been also found to predict fracture initiation conditions for solutions having a critical shear stress of two orders of magnitude lower than that required for melts.<sup>20,21</sup> Melt fracture is a gross distortion for which the whole of the molten polymer participates as it flows through the entry and the die. It would seem appropriate to use an average value of recoverable shear as a criterion for melt fracture instead of its value at the die wall. Consequently, if the recoverable shear strain hypothesis is valid, fracture initiation should occur at the same value of an average  $\overline{\sigma}_c$  independent of die geometry used, i.e.,

$$(\overline{\sigma}_c)_{\text{slit}} = (\overline{\sigma}_c)_{\text{cap}} = \overline{\sigma}_c = \text{const.}$$

The value of the constant will probably be different from polymer to polymer and a function of molecular weight distribution. A physically meaningful way to define an average recoverable shear is to use a flow-average value:

$$(\overline{\sigma})_{cap} = \frac{\int_{0}^{R} 2\pi r v(r) \sigma(r) dr}{\int_{0}^{R} 2\pi r v(r) dr}$$
for capillaries  
$$(\overline{\sigma})_{slit} = \frac{2 \int_{0}^{h/2} v(x) \overline{s}(x) dx}{2 \int_{0}^{h/2} v(x) dx}$$
for slits

Assuming a fully developed velocity profile for a power-law fluid flowing in a tube with circular cross section and  $\sigma(r) = J_0 \tau_w r/R$ , we have

$$\frac{(\overline{\sigma})_{\text{cap}}}{(\sigma_w)_{\text{cap}}} = \frac{2(1+3n)}{3(1+4n)}$$

Similarly, for slits,

$$\frac{(\overline{\sigma})_{\text{slit}}}{(\sigma_w)_{\text{slit}}} = \frac{1+2n}{2(1+3n)}$$

For a Newtonian fluid, the ratio of the flow-average recoverable shear to the recoverable shear to the wall is 8/15 for capillaries and 3/8 for slits. At critical conditions for melt fracture, we would have

$$\frac{(\sigma_{wc})_{\text{slit}}}{(\sigma_{wc})_{\text{cap}}} = \frac{4(1+3n)^2}{3(1+2n)(1+4n)}$$

This ratio has a value of 1.42 for a Newtonian fluid and is not very sensitive to the power-law index n. Calculated values for the materials studied are given in Table VII and are found to be in relatively good agreement with the experimental results of Tables IV-VI.

TABLE VII Calculated Critical Recoverable Shear Strain Ratios (at the Wall)

	Material	Power law index $n$	$(\sigma_{wc})_{\rm slit}/(\sigma_{wc})_{\rm cap}$			
	( Narrow MWD	0.41	1.38			
PS	$(M_w = 1.1 \times 10^5)$ Narrow NWD $(\overline{M}_w = 2.0 \times 10^5)$	0.29	1.37			
	HF-55	0.28	1.37			
	HF 42-1	0.51	1.39			
	Styron 456	0.39	1.38			
	/ DFDY-0701	0.46	1.39			
	DFDQ-0118	0.43	1.38			
	UFG-1	0.41	1.38			
LDPE	<b>DFDY-6005</b>	0.36	1.37			
	DYNH-5	0.41	1.38			
	Lupolen 2000H	0.435 a	1.38			
	(HD-211	0.568	1.39			
HDPE	<b>{ DMDJ 4309</b>	0.452	1.38			

<sup>a</sup>Supplied by G. Ehrmann.<sup>10</sup>

Using a flow-average recoverable shear (rather than its value at the die wall) as a criterion for melt fracture might be consistent with the observation that the initiation site for melt fracture is at the die entry. The flow-average recoverable shear strain is simply representative of the total amount of "straining" of the melt in shearing flow in the die and indirectly (through  $\dot{\gamma}$ ) is related to the amount of extensional deformation in the entry. It should also be noted that the early proposals<sup>22,23</sup> for the constancy of critical recoverable shear at fracture initiation conditions were based on  $\sigma_{\rm crit}$  estimates from entrance correction data. Such a quantity is more likely to correspond to an "average" value of  $\sigma$  than to a value estimated at the wall of a die.

# CONCLUSIONS

The objective of this paper was to compare the critical conditions for melt fracture initiation in capillaries and slits. The onset of gross distortion was relatively easy to identify when capillary dies were used. Three types of extrudate distortion were observed in extrusion through slit dies: (a) a midsection buckling at low shear stresses (much lower than  $(\tau_{wc})_{cap}$ ) which, in our opinion, is due to the restricting influence of the slit die edges; (b) a surface roughness which was very pronounced for LDPE ("ridge-like") but rather ill defined or absent for PS and HDPE: (c) a gross distortion which was characterized by the appearance of "ragged edges" and accompanied by an overall degradation of extrudate quality. It was assumed that the onset of spiral or wavy-type distortion in capillaries corresponds to the appearance of "ragged edges" in slits. Melt fracture in slits was found to initiate at higher wall shear stresses than in capillaries. It was proposed that an appropriate criterion for melt fracture is the flow-average value of the recoverable shear strain. Using this criterion, we obtained the ratio of critical recoverable shear strains at the wall  $(\sigma_{wc})_{slit}/(\sigma_{wc})_{cap}$  $\approx$  1.4, which is in relatively good agreement with experimental values based on shear stress and semiempirical expressions for shear compliance.

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