Polymer Bulletin 11, 223-228 (1984)

Polymer Bulletin

© Springer-Verlag 1984

Rheology

The Rheological Behavior of HDPE/LDPE Blends 4. Instability Phenomena in Capillary Flow

D. Acierno, F. P. La Mantia, D. Curto

Istituto di Ingegneria Chimica, University of Palermo, Italy

SUMMARY

Experimental data, relative to instability phenomena in capillary flow, have been collected for HDPE/LDPE blends already characterized from a rheological viewpoint. The data comprise morphological observations as well as the determination of critical parameters as a function of the blend composition and of the processing conditions.

INTRODUCTION

The high density polyethylene and the low density polyethylene show many differences with regard to both the morphology of the extruded samples when instability phenomena occur and to the phenomenon itself (DEN OTTER, 1970; VLACHOPOULOS and ALAM, 1972; WHITE, 1973; PETRIE and DENN, 1976).
It seemed thus very interesting to investigate the behaviour of the blends both from a rather fundamental viewpoint and from a more practical one. To this end data have been collected relative to samples already well characterized from a physico-chemical and a rheologicalviewpoint (LA MANTIA et al., 1982; CURTO et al., 1983; LA MANTIA et al., 1984). The data comprise morphological observations as well as the determination of the critical parameters as a function of the blend composition and of the processing conditions.

EXPERIMENTAL

The materials employed in this work were commercially available high density and low density polyethylenes manufactured and kindly supplied by Hoechst and Solvay (HDPE) and Montedison (LDPE). In the following the samples will be denoted as HDPE-X and LDPE-X with X equal to 1,2 or 3, increasing X corresponding to decreasing melt flow index (see TABLE 1). Three type of blends were prepared so as to cover the largest range of MFI ratios: HDPE-3/LDPE-1, HDPE-2/LDPE-2 and HDPE-1/LDPE-3. In each case blends were prepared with various weight percentages φ, of LDPE equal to 0, 20, 40, 60, 80 and 100% (i.e. various compositions). The blends were prepared by melt mixing the homopolymer parents in a laboratory mixer (Rheocord, Haake) at 210 °C and about 20 rpm. The mixing time was about 20 min, which was sufficient in all cases to obtain steady-

TABLE 1

Sample code	M.F.I.	Sample code	M.F.I.
HDPE-1	4.90	LDPE-1	3.90
HDPE-2	0.98	LDPE-2	0.74
HDPE-3	0.41	LDPE-3	0.07

state temperature and torque. The homopolymers were also subjected to the same procedure.

The rheological measurements were carried out with the aid of a constant rate capillary viscometer, Rheoscope 1000, manufactured by CEAST (Turin, Italy). The tests have been carried out at 160, 180 and 200 °C.

The critical shear rate has been considered as the shear rate at which the first irregularities, even only superficial, appear. This could be accurately done with the Rheoscope 1000 due to the continuous piston speed variation. Determinations of $\dot{\gamma}_c$ have been performed with different capillaries (D=1mm, L/D=5, 20, and 40).

RESULTS AND DISCUSSION

An important difference between high density and low density polyethylene is in the morphology of the extruded samples. It is well known for instance that the appearance of the HDPE samples changes strongly with the extrusion rate: at once the extrudate shows a mattness and then a screw-threadlike shape; the very regular spirals change into irregular ones and at higher shear rates the material is further distorted. The LDPE on the contrary shows since the beginning of the instability phenomenon an irregularly distorted extrudate; as expected the irregularities become gross and gross increasing the extrusion rate. The blends show an intermediate behaviour and FIG. 1 represents, as an example, photographs relative to the system HDPE-2/LDPE-2. A periodical swelling is shown by the HDPE at the critical shear rate while a distorted shape is shown by LDPE; for the blend with a 20% LDPE content distorted pieces alternate with swollen zones showing a screwlike surface; increasing the LDPE content the distorsions are more evident and the swollen zones are smaller; the blend with φ=80% shows distorsions similar to those of the pure LDPE but some regularities, on the long range, can be observed.

Also in the flow curves there is an effect of the blend composition. FIG. 2 represents for one of the systems (HDPE-3/LDPE-1) the shear stress vs the apparent shear rate. Indeed for the HDPE a pressure drop fluctuation corresponds to the beginning of the instabilities and the shear stress does not further increase with the shear rate while the flow curve relative to the LDPE shows only the continuous decrease in the slope. The behaviour of the blends is essentially similar to that of the low density polyethylene and in fact only for φ=20% a behaviour quantitatively similar to that of HDPE is observed.

From a practical viewpoint more quantitative data are necessary and FIG. 3 reports the apparent critical shear rate, at

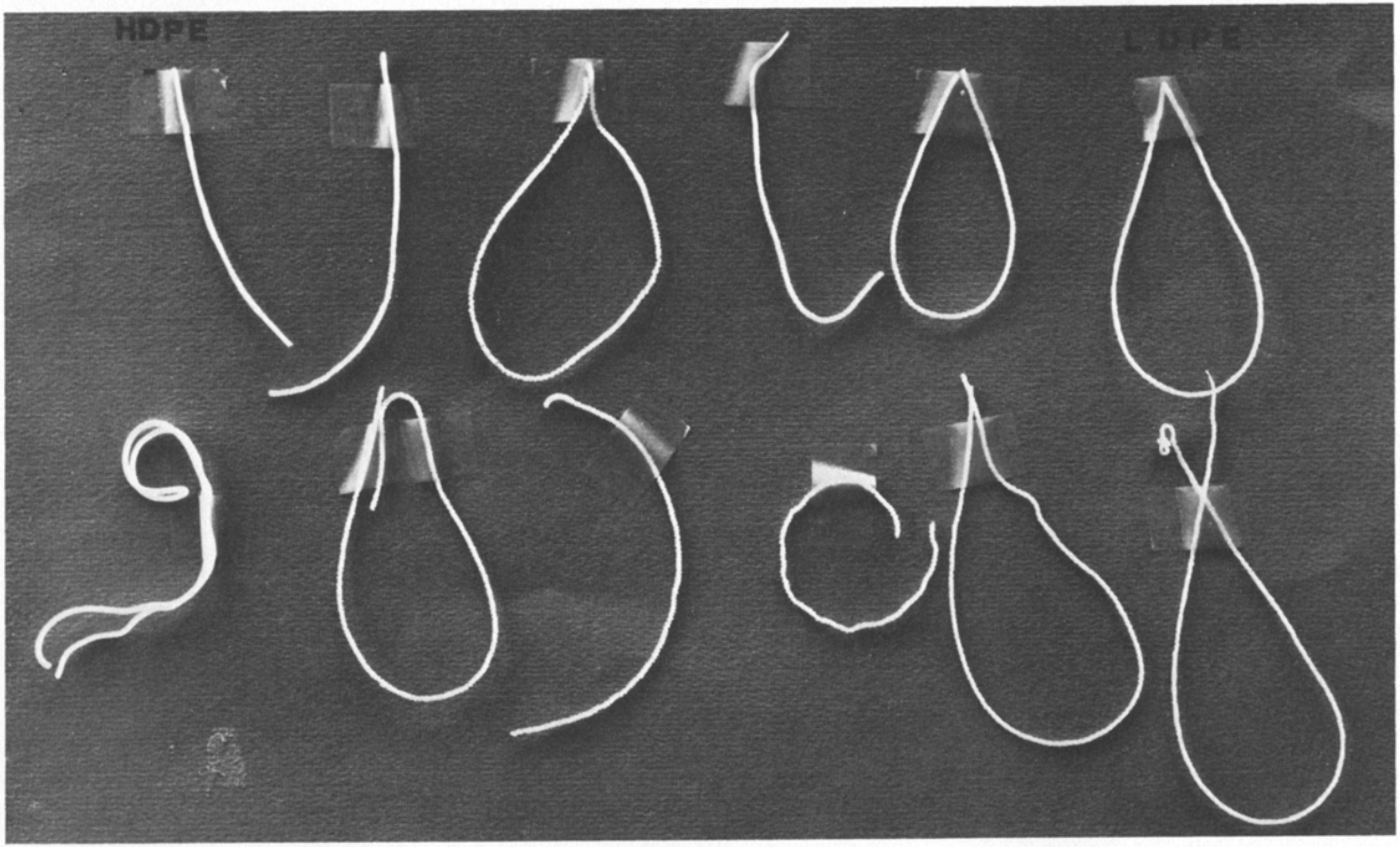

FIG.1 Extrudate of HDPE-2, LDPE-2 and their blends at φ 20,40,60 and 80%. T=180 °C; L/D=40
Top pictures refer to $\dot{\gamma}_c$, bottom pictures refer to $\dot{\gamma}=2\dot{\gamma}_c$

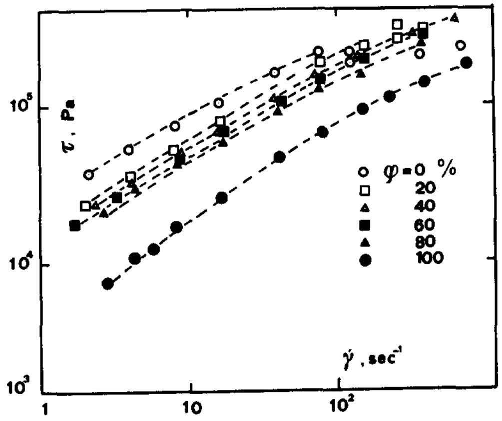

FIG.2 The shear stress as a function of the apparent shear rate for the HDPE-3/ LDPE-1 system at T=180 °C

one temperature and for two different length to diameter ratios as a function of the composition for all the systems. Two different behaviours can be observed: for the blends HDPE-1/LDPE-3 and HDPE-2/LDPE-2 the $\dot{\gamma}_c$ of the blends is, for any L/D, between those of the polymer parents; for the system HDPE-3/LDPE -1, i.e. the system for which the low density polyethylene shows in the considered conditions critical shear rate values similar or even larger than those of the high density to which it was blended, a pronunced minimum is present.

The effect of the capillary geometry (length to diameter ratio) is particularly interesting and indeed while $\dot{\gamma}_c$ decreases decreasing the L/D ratio for the HDPE, the opposite occurs for LDPE. The blends show an intermediate behaviour and a particu-

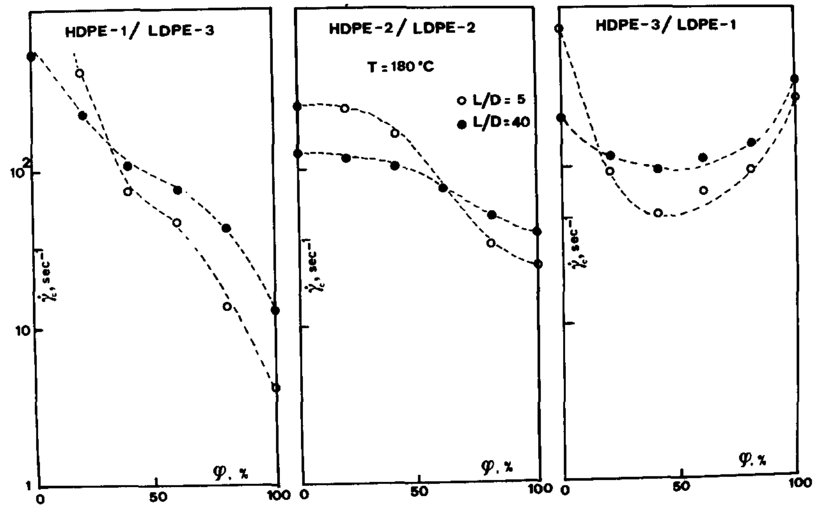

FIG.3 The apparent critical shear rate as a funcion of the blend composition

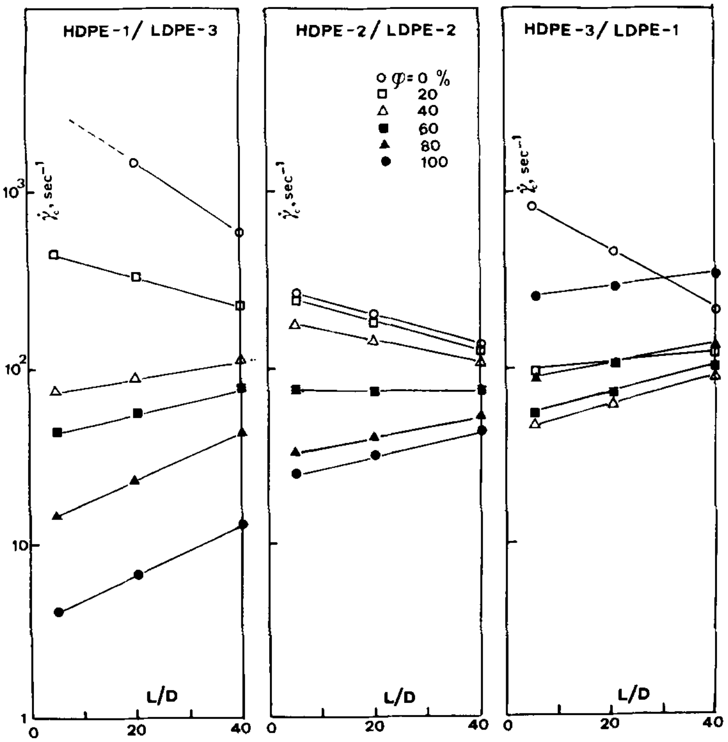

FIG.4 The apparent critical shear rate as a function of the length to diameter ratio

lar composition shows no dependence of the critical shear rate on the capillary length to diameter ratio. This particular composition is not however the same for the three types of blends. This feature is well evidenced in FIG.4, where the critical shear rate is plotted as a function of L/D for all the blends. For the system HDPE-2/LDPE-2 at φ=60% the critical shear rate is independent from the L/D ratio; the blends richer in HDPE show a behaviour similar to that of HDPE,while the contrary occurs for the blends rich in LDPE. For the other two systems the transition occurs with smaller content of LDPE, about 20%. FIG.5 shows the effect of the temperature on the critical shear rate: the $\dot{\gamma}_c$ at L/D=40 is plotted for the system HDPE-2/LDPE-2 as a function of the inverse of the absolute temperature. It is well evident that, increasing the content of LDPE, $\dot{\gamma}_c$ depends more strongly on the temperature. This feature is very similar to the effect of the temperature on the viscosity;and indeed the activation energy of the viscous flow for the LDPE is about twice that of the HDPE.

Finally the critical shear stress, reported in FIG.6 for all the systems, is less sensitive to the composition and is always for the blends in between those of the high density and of the low density polyethylene.

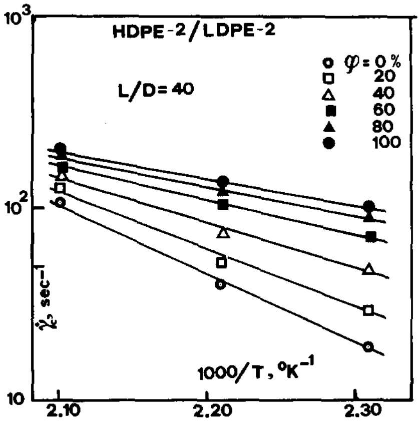

FIG.5 The apparent critical shear rate as a function of the inverse of temperature for the HDPE-2/LDPE-2 system

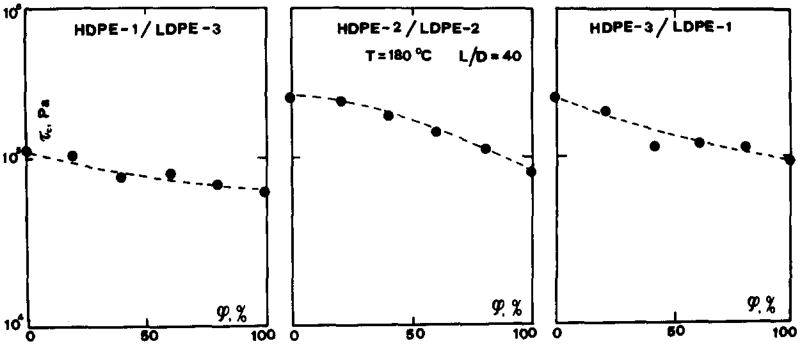

FIG.6 The critical shear stress as a function of the blend composition

ACKNOWLEDGMENT

This work has been carried out with the financial support of "Progetto Finalizzato del CNR Chimica Fine e Secondaria".

REFERENCES

CURTO,D., LA MANTIA,F.P., ACIERNO,D.: Rheol Acta, 22,197 (1983)
DEN OTTER,J.L.: Plastics Polymers, 38, 155 (1970)
LA MANTIA,F.P., ACIERNO,D., CURTO,D.:Rheol.Acta, 21,452 (1982)
LA MANTIA,F.P., CURTO,D., ACIERNO,D.: Acta Polymerica, in press
PETRIE,C.J.S. and DENN,M.M.:A.I.Ch.E.J., 22,209 (1976)
VLACHOPOULOS,J. and ALAM,M.: Polym. Eng. Sci., 12,184 (1972)
WHITE,J.L.: Polym. Symp., 20,155 (1973)

Accepted December 14, 1983