

EVALUATION OF MELT FLOW INSTABILITIES OF HIGH-DENSITY POLY-ETHYLENES VIA AN OPTIMISED METHOD FOR DETECTION AND ANALYSIS OF THE PRESSURE FLUCTUATIONS IN CAPILLARY RHEOMETRY

SUSANA FILIPE^{1,2,+}, ALFONS BECKER¹, VITOR C. BARROSO^{1,2}, MANFRED WILHELM^{1,2*}

¹ Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany

² Institute for Chemical Technology and Polymer Chemistry, Karlsruhe Institute of Technology (KIT), Engesserstrasse 18, 76128 Karlsruhe, Germany

+ current address: Borealis GmbH, St. Peter Strasse 25, 4021, Linz, Austria

* Email: wilhelm@polymer.uni-karlsruhe.de

Fax: +49.721.6083153

Received: 26.6.2008, Final version: 12.12.2008

ABSTRACT:

An optimised method for the detection and analysis of the time dependent pressure associated with the development of melt flow instabilities during extrusion through a capillary die was developed and validated. The magnitude and frequency of the developed quasi-periodic distortions, as well as the pressure profiles along the die length, were found to depend on the MWD, topology, melt elasticity and uniaxial extensional flow properties. Both the onset and magnitude of strain hardening in uniaxial extension appear to be related to the onset for the development of melt flow instabilities under capillary flow. For a better understanding of the role of the extensional properties (namely that of a purely elastic instability) the Hencky strain to failure was also determined and correlated to the observed flow instabilities. Time resolution of the capillary rheometer was improved by a factor of 1000, pressure resolution by a factor of 100 compared to the original set-up.

ZUSAMMENFASSUNG:

Eine neuartige optimierte Methode zur Erfassung und Analyse des zeitabhängigen Drucks wurde entwickelt, um die entstehenden Fließinstabilitäten durch Extrusion mit einer Kapillardüse bezüglich der Zeit und Intensität zu quantifizieren. Sowohl die Größenordnung und die Frequenz der entstehenden quasi-periodischen Formänderungen, als auch die Druckprofile entlang der Düsenlänge, sind von der Molmassenverteilung (MWD), der Topologie, der Elastizität der Schmelze und den Dehnfließeigenschaften abhängig. Die Entstehung wie auch die Größenordnung der Dehnverfestigung beeinflussen die Fließinstabilitäten der Schmelze während der Kapillarextrusion. Die Hencky-Dehnung, bei der die Probe versagt wurde bestimmt, und mit den beobachteten Fließinstabilitäten korreliert, um den Einfluß der Dehneigenschaften (vor allem der rein elastischen Instabilität) zu verstehen. Die Zeitauflösung wurde dabei um den Faktor 1000, die Druckauflösung um den Faktor 100 verbessert.

RÉSUMÉ:

Une méthode optimisée pour la détection et l'analyse de la dépendance temporelle de la pression associée avec le développement des instabilités d'écoulement d'un fondu lors de l'extrusion dans une filière capillaire a été développée et validée. L'amplitude et la fréquence des distorsions quasi-périodiques développées, ainsi que les profils de pression le long de la filière, se sont avérés dépendre de la distribution de masse moléculaire MWD, de la topologie, de l'élasticité du fondu et des propriétés d'écoulement en extension uni axiale. L'émergence et l'amplitude du durcissement à la déformation semblent être reliées à l'émergence du développement des instabilités d'écoulement du fondu dans un écoulement capillaire. Afin de mieux comprendre le rôle des propriétés extensionnelles (c'est-à-dire celui d'une instabilité purement élastique), la déformation de Henky à la rupture a été également déterminée et corrélée avec les instabilités d'écoulement observées.

KEY WORDS: Polyethylenes, stick-slip, sharkskin, Fourier transformation, molecular topology, capillary rheology, uniaxial extensional flow

© Appl. Rheol. 19 (2009) 23345

This is an extract of the complete reprint-pdf, available at the Applied Rheology website

<http://www.appliedrheology.org>

Applied Rheology

Volume 19 · Issue 2

<http://www.appliedrheology.org>

The results from this work point towards the conclusion that the onset shear rate at which melt flow instabilities occur correlates with the extensional flow properties, particularly the viscosity (or stress) growth. The critical condition for the onset of melt flow instabilities shifts to higher shear rates for materials showing failure at higher Hencky strain (such as LCB3) or materials with a lower M_w (such as LCB1). The development of melt flow instabilities such as stick-slip and helicoidal defect are highly influenced by the extensional properties of the material. The use of this non-conventional set-up should be extended in the future to the characterisation of melt flow instabilities on materials with a linear structure and a narrow molecular weight distribution. Such analysis will certainly help to get further insights on defects arising at the die, e.g. shark-skin.

ACKNOWLEDGEMENTS

An acknowledgement is due to Dr. A. Göttfert, Dr. J. Sunder, and Dipl.-Ing. U. Bäuerle from the company GÖTTFERT GmbH, as well as Dipl.-Ing. Meyer and Dipl.-Ing. Stahl from KISTLER, for the intensive discussions and support. The authors would also like to thank Dipl.-Ing. I. Naue (TU Darmstadt, Fachbereich Maschinenbau) and to Dr. I. Vittorias for their support.

REFERENCES

- [1] Larson RG: Instabilities in viscoelastic flow, *Rheol. Acta* 31 (1992) 213-263.
- [2] Miller E, Rothstein JP: Control of the sharkskin instability in the extrusion of polymer melts using induced temperature gradients, *Rheol. Acta* 44 (2004) 160-173.
- [3] Münstedt H, Schmidt M, Wassner E: Stick and slip phenomena during extrusion of polyethylene melts investigated by laser-Doppler velocimetry, *J. Rheol.* 44 (2000) 413-427.
- [4] Carreras ES, El Kissi N, Piau JM, Toussaint F, Nigen S: Pressure effects on the viscosity and flow stability of polyethylene melts during extrusion, *Rheol. Acta* 45 (2006) 209-222.
- [5] Deepak C, Rosenblatt C, Wang S-Q: Molecular character of sharkskin phenomenon in metallocene linear low density polyethylenes, *Macromol. Chem. Phys.* 199 (1998) 2113-2118.
- [6] Denn MM: Extrusion instabilities and wall slip, *Annu. Rev. Fluid Mech.* 33 (2001) 265-287.
- [7] Doerpinghaus PJ, Baird DG: Comparison of the melt fracture behaviour of metallocene and conventional polyethylenes, *Rheol. Acta* 42 (2003) 544-556.
- [8] Fernandez M, Vega JF, Santamaria A, Muñoz-Escalona A, Lafuente P: The effect of chain architecture on "shark-skin" of metallocene polyethylenes, *Macromol. Rapid Commun.* 21 (2000) 973-978.
- [9] Robert L, Demay Y, Vergnes B: Stick-slip flow of high density polyethylene in a transparent slit die investigated by laser Doppler velocimetry, *Rheol. Acta*, 43 (2004) 89-98
- [10] Robert L: Instabilité oscillante de polyethylenes linéaires: observations et interprétations, Ph. D. thesis, Nice Sophia-Antipolis (2001).
- [11] Wang S-Q: Molecular transitions and dynamics at polymer / wall interfaces: origin of flow instabilities and wall slip, *Adv. Polym. Sci.* 138 (1999) 228-275.
- [12] Hatzikiriakos SG, Migler KB: Polymer Processing Instabilities: Control and Understanding, First Edition, CRC Press (2004).
- [13] Achilleos E, Georgiou GC, Hatzikiriakos SG: On numerical simulation of polymer extrusion instabilities, *Appl. Rheol.* 12 (2002) 88-104.
- [14] Hristov V, Vlachopoulos J: A study of entrance pressure loss in filled polymer melts, *Appl. Rheol.* 17 (2007) 57191.
- [15] Zatloukal M, Vlcek J, Slanik A, Lengalova A, Simonik J: Experimental and numerical investigation into metallocene polymer melt flow in film blowing dies, *Appl. Rheol.* 12 (2002) 126-132.
- [16] Nithi-Uthai N, Manas-Zloczower I: Numerical simulation of sharkskin phenomena in polymer melts, *Appl. Rheol.* 13 (2003) 79-86.
- [17] Joshi YM: Studies on wall-slip in entangled polymeric liquids, *Appl. Rheol.* 11 (2001) 277-280.
- [18] El Kissi N, Piau JM, Toussaint F: Sharkskin and cracking of polymer melt extrudates, *J. Non-Newtonian Fluid Mech.* 68 (1997) 271-290.
- [19] El Kissi N, Piau JM, Adhesion of linear low density polyethylene for flow regimes with sharkskin, *J. Rheol.* 38 (1994) 1447-1463.
- [20] El Kissi N, Piau JM: The different flow regimes of entangled polydimethylsiloxane polymers: macroscopic slip at the wall, *J. Non-Newtonian Fluid Mech.* 37 (1990) 55-94.
- [21] Doelder J, Koopmans R, Dees M, Mangnus M: Pressure oscillations and periodic extrudate distortions of long-chain branched polyolefins, *J. Rheol.* 49 (2005) 113-126.
- [22] Wang S-Q, Drda PA, Inn Y-W: Exploring molecular origins of sharkskin, partial slip, and slope change in flow curves of linear low density polyethylene, *J. Rheol.* 40 (1996) 875-898.
- [23] Hassel D, Mackley M, Harlen OG: Effect of branching on polyethylene melts within Cross-Slot flow, in Proceedings of the Annual European Rheology Conference 2007, page 167.

This is an extract of the complete reprint-pdf, available at the Applied Rheology website
<http://www.appliedrheology.org>

- [24] Wilhelm M, Reinheimer P, Ortseifer M: High sensitivity Fourier-transform rheology, *Rheol. Acta* 38 (1999) 349-356.
- [25] Wilhelm M: Fourier Transform-Rheology, *Macromol. Mat. Eng.* 287 (2002) 83-105.
- [26] van Dusschoten D, Wilhelm M: Increased torque transducer sensitivity via oversampling, *Rheol. Acta* 40 (2001) 395-399.
- [27] Hilliou L, van Dusschoten D, Wilhelm M, Bührin H, Rodger ER: Increasing the torque force transducer sensitivity of an RPA 2000 by a factor of 5-10 via advanced data acquisition, *Rubber Chem. Technol.* 77 (2004) 192-200.
- [28] Klein C, Venema P, Sagis L, van Dusschoten D, Wilhelm M, Spiess HW, van der Linden E, Rogers SS, Donald AM: Rheo-optical measurements using Fast Fourier Transform and oversampling, *Appl. Rheol.* 17 (2007) 45210.
- [29] Rutgers R, Mackley M: The correlation of experimental surface extrusion instabilities with numerically predicted exit surface stress concentrations and melt strength for low linear density polyethylene, *J. Rheol.* 44 (2000) 1319-1334.
- [30] Migler KB, Son Y, Qiao F, Flynn K: Extensional deformation, cohesive failure, and boundary conditions during sharkskin melt fracture, *J. Rheol.* 46 (2002) 383-400.
- [31] Pérez R, Rojo E, Fernández M, Leal V, LaFuente P, Santamaría, A: Basic and applied rheology of m-LLDPE/LDPE blends: miscibility and processing features, *Polymer* 46 (2005) 8045-8053.
- [32] Pollard M, Klimke K, Graf R, Spiess HW, Sperber O, Piel C, Kaminsky W: Observation of chain branching in polyethylenes in solid state and melt via ^{13}C NMR spectroscopy and melt relaxation time measurements, *Macromolecules* 37 (2004) 813-825.
- [33] Klimke K, Parkinson M, Piel C, Kaminsky W, Spiess HW, Wilhelm M: Optimisation and application of polyolefin branch quantification by melt-state ^{13}C NMR spectroscopy, *Macromol. Chem. Phys.* 207 (2006) 382-39.
- [34] Filipe S, Vittorias I, Wilhelm M: Experimental correlation between mechanical non-linearity in LAOS flow and capillary flow instabilities for linear and branched commercial polyethylenes, *Macromol. Mat. Eng.* 293 (2008) 57-65.
- [35] Wagner MH, Bastian H, Hachmann P, Meissner J, Kurzbeck S, Münstedt H, Langouche F: The strain-hardening behaviour of linear and long-chain-branched polyolefin melts in extensional flows, *Rheol. Acta* 39 (2000) 97-109.
- [36] Gabriel C, Münstedt H: Strain hardening of various polyolefins in uniaxial elongational flow, *J. Rheol.* 47 (2003) 619-630.
- [37] Sentmanat M, Wang BN, McKinley GH: Measuring the transient extensional rheology of polyethylene melts using the SER universal testing platform, *J. Rheol.* 49 (2005) 585-606.
- [38] Wood-Adams PM, Dealy JM, de Groot AW, Redwine OD: Effect of molecular structure on the linear viscoelastic behavior of polyethylene, *Macromolecules* 33 (2000) 7489-7499.
- [39] Züller B, Linster JJ, Meissner J, Hürlimann HP: Deformation hardening and thinning in both elongation and shear of a low density polyethylene melt, *J. Rheol.* 31 (1987) 583-598.
- [40] Dealy JM: Do polymeric liquids exhibit strain hardening?, *J. Rheol.* 34 (1990) 1133-1147.
- [41] Schweizer T: The uniaxial elongational rheometer RME – six years of experience, *Rheol. Acta* 39 (2000) 428-443.
- [42] Dealy JM, Larson RG: Structure and Rheology of Molten Polymers, First Edition, Hanser Publishers, Munich, Germany (2006), pages 213, 352, 381-383.
- [43] Considère M: Mémoire sur l'emploi du fer et de l'acier dans les constructions, *Annales des Ponts et Chausées* 9 (1885) 574-605.
- [44] Wang Y, Boukany P, Wang S-Q, Wang X: Elastic breakup in uniaxial extension of entangled polymer melts, *Phys. Rev. Lett.* 99 (2007) 237801.
- [45] McKinley GH, Hassager O: The Considère condition and rapid stretching of linear and branched polymer melts, *J. Rheol.* 43 (1999) 1195-1212.
- [46] Barroso VC, Maia JM: Influence of long-chain branching on the rheological behavior of polyethylene in shear and extensional flow, *Polym. Eng. Sci.* 45 (2005) 984-997.



This is an extract of the complete reprint-pdf, available at the Applied Rheology website
<http://www.appliedrheology.org>

This is an extract of the complete reprint-pdf, available at the Applied Rheology website
<http://www.appliedrheology.org>

Applied Rheology
 Volume 19 · Issue 2

23945-14