

MIXING IN EXTENSIONAL FLOW FIELD

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ABSTRACT

Mixing is the most important operation in polymer processing. Uniformity of the molecular weight, degree of entanglement, temperature, and composition, is the prerequisite for good performance. Mixing of molten polymers is always within the laminar (as opposed to turbulent) flow regime. This paper presents an overview of the effect of flow type on mixing. The distributive mixing is best modeled considering either laminar or chaotic mixing. Here the system is "passive", i.e. the rheological properties are identical and the interface is "invisible". Furthermore, the effects of flow and morphology are "decoupled". By contrast, the dispersive mixing is best modeled by means of microrheology, complemented by coalescence. Both models – the laminar mixing and the microrheology – show the unambiguous superiority of the extensional flows for dispersive and distributive mixing, in particular, when the viscosity ratio of two principal components is large. However this superiority is balanced by practical difficulties in generating a flow that may ascertain a high level of extension.

Results of simulation and model experiments on drop deformability in converging and diverging (c-d) flow were used to develop a simple and inexpensive mixer capable of generating a high level of extension that lead to enhanced mixing capability. In consequence, an extensional flow mixer (EFM) was designed. In this device, a liquid mixture is repeatedly exposed to extensional flow fields and semi-quiescent zones. The c-d channels are of progressively increasing intensity. To reduce the pressure drop, and to prevent blockage, slit restrictions are used. The EFM (as well as its newer, dynamic version – DEFM) is fully adjustable, general-purpose mixers. Several its versions have been used for: polymer blending, incorporation of elastomers into resins, and dispersion of high viscosity polymers or "gel particles". For most mixing applications a single-screw extruder equipped with either EFM or DEFM performed at least as well as a co-rotating, intermeshing twin-screw extruder, with high efficiency screw configuration.

ZUSAMMENFASSUNG

Das Mischen ist eine der wichtigsten Operationen in der Kunststoffverarbeitung. Einheitlichkeit der Molmasse, Grad der Verschlaufungen, Temperatur und Komposition sind die Voraussetzungen für gute Eigenschaftsprofile der Mischungen. Das Mischen von Polymerschmelzen erfolgt grundsätzlich im laminaren (im Gegensatz zum turbulenten) Strömungsbereich. Dieser Beitrag präsentiert einen Überblick über den Einfluß der Strömungsform auf das Mischen und das Mischergebnis. Das distributive Mischen wird am besten durch Annahme von laminarem oder chaotischem Mischvorgängen modelliert. Das System ist in diesem Fall „passiv“, d.h. die rheologischen Eigenschaften sind identisch und die Wechselwirkungen mit der Phasengrenze sind „unsichtbar“. Ferner sind die Einflüsse von Strömung und Morphologie entkoppelt. Das dispersive Mischen hingegen wird am besten durch mikrorheologische Eigenschaften, vervollständigt durch Koaleszenzvorgänge, modelliert. Beide Modelle, das laminare Mischen und die Mikrorheologie, belegen den eindeutigen Vorteil einer Dehnströmung für disperses und distributives Mischen, insbesondere wenn das Viskositätsverhältnis der beiden Komponenten groß ist. Dieser Vorteil wird jedoch durch die praktischen Schwierigkeiten bei der Generierung eines Strömungsfeldes mit einem hohen Dehnanteil begrenzt.

Die Ergebnisse von Simulationen und Modellexperimenten zur Deformierbarkeit von Tropfen in konvergenter und divergenter (c – d) Strömung wurden für die Entwicklung eines einfachen und kostengünstigen Mischers verwendet, wobei der Mischer für die Generierung eines hohen Dehnströmungsanteiles, die zu verbesserten Mischeigenschaften führt, geeignet ist. Das Ergebnis dieser Überlegung floß in die Konstruktion eines Dehnströmungsmischer ein (Extensional Flow Mixer). In dieser Vorrichtung ist die Flüssigkeitsmischung abwechselnden Dehnströmungsfeldern und Ruhezeiten ausgesetzt wobei die c – d-Lastungszonen von zunehmender Intensität sind. Um den Druckverlust zu reduzieren und das Blockieren der Strömung zu verhindern, wurden Schlitzbegrenzungen verwendet. Der Dehnungsmischer (EFM) und eine weiterentwickelte dynamische Version (DEFM) sind voll verstellbare Allzweckmischer. Mehrere Versionen dieser Mischer wurden für die Herstellung von Polymermischungen, für die Einarbeitung von Elastomeren in thermoplastische Kunststoffe und für die Dispergierung von hochviskosen Polymeren oder Gelpartikeln eingesetzt. Für die meisten Compoundieraufgaben erreichen Einwellenextruder ausgerüstet mit einem EFM- oder einem DEFM-Mischer eine ähnliche Effizienz wie ein gleichsinnig drehender, dichtkämmender Doppelwellenextruder mit hocheffizienter Schneckenkonfiguration.

RÉSUMÉ

Mélanger est l'action la plus importante dans la mise en oeuvre des polymères. L'uniformité dans le poids moléculaire, dans le degré d'enchevêtrement, dans la température et la composition, est une condition préalable pour l'obtention de bonnes performances. Le mélange de polymères fondus est toujours considéré dans le régime d'écoulement laminaire (par opposition au régime d'écoulement turbulent). Cet article présente une revue de l'effet du type d'écoulement sur

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Table 5:
The systems used in the
model studies [19].

Matrix fluid	Drop Fluid	η_m (Pas)	η_d (Pas)	λ	ν_{12} (mN/m)
PDMS ¹	PB (Newtonian)	60	530	8.83	3.65
PDMS	PB (Newtonian)	60	180	3	3.65
PDMS	1% PAM in H ₂ O	60	600	10	12.98
Corn Syrup	0.075% HMW PS in PS-DOP 28/72 (Boger fluid)	6.0	13.5	2.25	5.87

Notes: ¹ PDMS — polydimethylsiloxane, PB — polybutene-1, PAM — polyacrylamide, PS — polystyrene, HMW PS — high molecular weight polystyrene, DOP — di(ethylhexyl)phthalate.

ing, indicating that in convergent flow of polymer melts the moderate SH will be compensated by pseudoplasticity.

2.4 EFM MIXING EFFICIENCY

Performance of EFM attached to a single-screw extruder (SSE) was examined in reference to the results obtained using a co-rotating, intermeshing twin-screw extruder (TSE) equipped with the recommended by the manufacturer high dispersion screws. Four types of mixing were studied: dispersion of a viscous polymeric blend component in at least four times less viscous polymer matrix, impact-modification of engineering resins, elimination of gel particles from reactor powders, and homogenization of new metallocene polyolefin mixtures having very large ratios of the melt flow index. The results demonstrated that compounding in SSE+EFM resulted in finer dispersion of HDPE in PS and *vice-versa*, than in a TSE. Similarly, the impact properties of EPR/PP blends prepared in SSE+EFM were better than of the same compositions mixed in TSE. The SSE+EFM mixing unit was also superior to TSE as far as dissolution of very high molecular weight fractions (polymeric gel particles) was concerned [22-23].

As an example, the mechanical properties of three PET blends with metallocene PE (mPE) are listed in Table 6. These blends (with or without a compatibilizer) were prepared using either a 38 mm SSE+EFM (at 2000 psi or 14 MPa) or two compounding TSE (from different manufacturers).

The SEM images of these blends indicated similar morphology from these three systems, but for some blends SSE+EFM gave a finer dispersion. The tabulated data clearly indicate that on average SSE+EFM outperformed either TSE machine. This conclusion is particularly pleasing since the cost of the former system is about six times lower \bar{n} for blending SSE+EFM system offers a serious alternative to the expensive TSE.

3 SUMMARY

Majority of commercial polymer blends is melt-blended, mostly in TSE, less frequently in SSE, and rarely in batch mixer or other specialty mixing devices. SSE is being used for the preparation of blends that either do not require compatibilization, or are compatibilized by addition of non-reactive compatibilizers. For both applications, SSE is frequently supplemented by additional mixing devices, viz. mixing screws, add-ons, extensional flow mixer, etc.

When selecting the mixing strategy one must consider the thermo-mechanical degradability of the blends ingredients (including the compatibilizer!), the time scale of the relaxation processes, as well as the stress-induced coalescence. Some polymers, *e.g.* PP, are sensitive to degradation. For this reason better performance may be obtained from PP/EPR blends using low dispersing energy, just sufficient to adequately disperse the elastomer without inducing too extensive degradation of the matrix.

Table 6:
Modulus, E, maximum
strain at break, ϵ , and
notched Izod impact
strength at room
temperature (NIRT) for
PET/mPe blends.

MPE (WT%)	E (GPa)		ϵ (%)		NIRT (J/m)	
	TSE	SSE+EFM	TSE	SSE+EFM	TSE	SSE+EFM
AMORPHOUS PET						
10	1.98±0.02	2.12±0.05	82±21	53±14	39.3	50.4
20	1.91±0.46	1.68±0.10	68±21	46±26	55.1	54.4
20+compat.	1.47±0.02	1.85±0.52	38.0±6	37.0±3	76.8	81.5
CRYSTALLINE PET						
10	2.44±0.09	2.36±0.11	9.5±0.7	13.3±2.7	29.0	35.3
20	2.09±0.36	1.85±0.13	17.5±3.6	14.5±2.6	43.3	42.3
20+compat.	2.07±0.28	2.04±0.20	13.1±2.7	12.7±2.0	61.5	58.5

One of the most efficient mechanisms of mixing is development of fibrils that upon reduction of stress disintegrate by the capillarity mechanism into thousands of micro-droplets. This mechanism is greatly facilitated by the extensional flow field.

For homogenization and mixing the newly developed Extensional Flow Mixer (EFM and DEFM) was found to offer inexpensive alternative to TSE. EFM provides good mixing at relatively low pressure loss and under conditions that do not cause extensive degradation to polymers. The mixer provided satisfactory results for homogenization (viz. elimination of gel particles or production of homogenous melt from PE fractions with melt index ratio up to 50,000), good dispersion of one polymer in another in immiscible blends with or without compatibilizer.

- [19] Bourry, D., Godbille, F., Khayat, R. E., Luciani, A., Picot, J., and Utracki, L. A., *Polym. Eng. Sci.*, 39, 1072 (1999).
- [20] Khayat, R. E., Luciani, A., and Utracki, L. A., in *Boundary Elements*, 8, 515 (1996); *Eng. Analysis with Boundary Elem.*, 19(4), 279 (1997).
- [21] Khayat, R. E., Luciani, A., and Utracki, L. A., and Goulet, J. F., in *Boundary Elements*, M. Marchetti, C. A. Brebbia and M. H. Aliabadi, Eds., *Computational Mechanics Publications*, 533 (1997).
- [22] Utracki, L. A., and Luciani, A., *Intl. Plast. Eng. Technol.*, 2, 37 (1996).
- [23] Luciani, A., and Utracki, L. A., *Intl. Polymer Process.*, 11, 299 (1996).



REFERENCES

- [1] Utracki, L. A., Ed., *Encyclopaedic Dictionary of Commercial Polymer Blends*, ChemTec, Toronto (1994).
- [2] Utracki, L. A., *Commercial Polymer Blends*, Chapman and Hall, London (1997).
- [3] Utracki, L. A., Ed., *Polymer Blends Handbook*, Kluwer Academic Press, Dordrecht (1999).
- [4] Ng, K. Y., and Erwin, L., *Polym. Eng. Sci.*, 21, 212 (1981).
- [5] Erwin, L., in *Mixing in Polymer Processing*, C. Rauwendaal, Ed., Marcel Dekker, New York (1991).
- [6] Poitou, A., PhD thesis, ...cole des Mines de Paris (1988).
- [7] Taylor, G. I., *Proc. Roy. Soc.*, (London), A138, 41 (1932); *ibid.*, A146, 501 (1934).
- [8] Tomotika, S. *Proc. Roy. Soc.*, A150, 322 (1935); *ibid.*, A153, 302 (1936).
- [9] Luciani, A., Champagne, M. F., and Utracki, L. A., *Polym. Networks Blends*, 6, 41, 51 (1996); *J. Polym. Sci. B, Polym. Phys. Ed.*, 35, 1393 (1997).
- [10] Oldroyd, J. C., *Proc. Roy. Soc.*, A232, (1955) 567.
- [11] Elmendorp, J. J., *Polym. Eng. Sci.*, 26, 418 (1986).
- [12] Utracki, L. A., *J. Coll. Interf. Sci.*, 42, 185 (1973); *Polym. Networks Blends*, 1, 61 (1991).
- [13] Spiegelberg, S. H., Ables, D. C., and McKinley, G. H., *J. Non-Newtonian Fluid Mech.*, 64, 229 (1996).
- [14] Larson, R. G., *Constitutive Equations for Polymer Melts and Solutions*, Butterworths, Boston (1988).
- [15] Lindt, J. T., and Ghosh, A. K., *Polym. Eng. Sci.*, 32, 1802 (1992).
- [16] Nguyen, X. Q., and Utracki, L. A., U. S. Pat., 5,451,106, 19 Sept. 1995, Appl. 08 Aug. 1984, to National Research Council of Canada, Ottawa, Canada.
- [17] Utracki, L. A. and Luciani, A., Extensional flow mixer, Canadian Pat. Appl., # 2,217,374, 29.09.1997; to National Research Council of Canada, Ottawa, Canada.
- [18] Utracki, L. A., Bourry, D., and Luciani, A., Extensional flow mixer (EFM-3 & DEFM), PCT Pat. Appl., #WO 99/16540, 08.04.1999, Appl. 29.09.1998, to National Research Council of Canada, Ottawa, Canada.