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An Empirical Model for the Melt Viscosity of Polymer Blends

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Summary

On the basis of experimental data for blends of polyethylene with different polymers an empirical equation is proposed to describe the dependence of melt viscosity of blends on component viscosities and composition. The model ensures the continuity of viscosity vs.composition curves throughout the whole composition range, the possibility of obtaining extremum values higher or lower than the viscosities of components, allows the calculation of flow curves of blends from the flow curves of components and their volume fractions.

Introduction

Establishing mathematical models for the quantitative correlation of rheological properties of polymer blends with composition has been a major preoccupation throughout the past ten – fifteen years. Solving this problem is important from a theoretical viewpoint since the models must take into account and subsequently explain the way blend properties are determined by component properties (such as chemical nature, molecular weight and molecular weight distribution, chain structure – tacticity, number, length and distribution of branches), as well as by phase structure, morphology and inter – phase adhesion. Such models are also important from a technological point of view since they would allow the direct estimation of rheological properties of blends of any composition on the basis of properties of pure components, and the proper selection of required composition. The task of modelling such a large number of factors whose effects are practically impossible to separate and evaluate individually is difficult and has not been accomplished as yet, especially for the melts. Several models have been proposed which apply to blends consisting of narrow fractions of different molecular weights of the same polymer. From among them are

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the Ninomiya blending law (NINOMIYA 1959, NINOMIYA 1962), the BMEO quadratic law (BOGUE et al.1970, MASUDA et al.1973),higher order blending laws (KURATA et al.1974) and the models advanced by Hong, Soong and Shen (HANSEN et al.1976,HONG et al.1977a,b,SOONG et al. 1979).Even for this narrow field of application,such models take intricate mathematical forms and their use require complex experimental data. Most blending laws employed for the viscosity of polymer blends in the melt are empirical and semiempirical equations,some of them initially developed for low molecular fluid mixtures or for suspensions and very few of them were specially derived for molten polymer blends.Some models which consider the component that forms the disperse phase give discontinuous viscosity vs. composition curves (UEMURA and TAKAYANAGI 1966).All equations known,no matter what the dependence of viscosity on composition they assume,yield blend viscosities ranging between values for pure components.In many instances,however,viscosities of blends take values either higher or lower than those of components (HAN and YU 1971,HAN et al.1973,ABLAZOVA et al.1975,DOBRESCU and COBZARU 1978,DOBRESCU 1980).The problem was to develop an equation to cover these cases as well.This empirical equation was intended to satisfy the following conditions:

- the continuity of viscosity vs.composition curve over the whole composition range ;
- the possibility of obtaining extremum values ;
- the absence of constraints as to nature and shape of disperse phase ;
- the possibility to calculate parameters only on the basis of steady state flow curves of components ;
- the possibility of utilizing the model to calculate the flow curves of blends of various compositions using the flow curves of components ;
- the possibility of utilizing the model for different types of blends.

New Model. Evaluation and Conclusions.

In a previous paper (DOBRESCU 1980), on the basis of the analysis of viscosity – composition dependence for polyethylene blends as well as for other blends, the following equation has been proposed :

$$\lg \eta = v_1 \lg \eta_1 + \mathfrak{I} v_1 v_2 + v_2 \lg \eta_2 \qquad (1)$$

where η, η_1 and η_2 are the viscosities at constant shear stress, or the

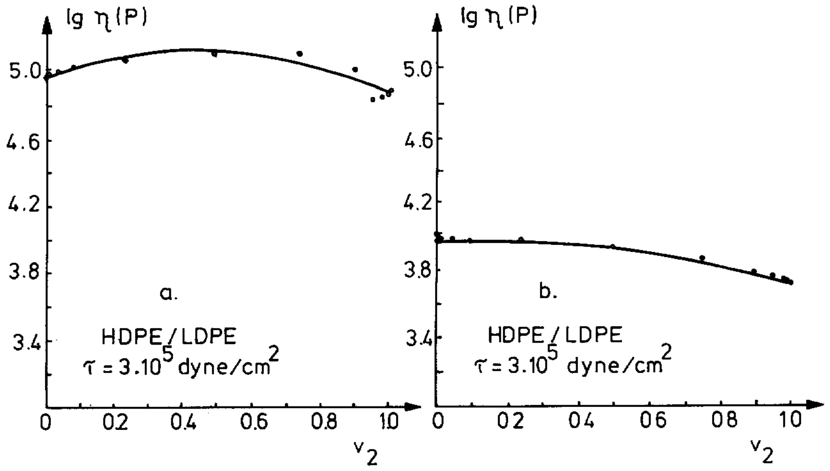

Figure 1. Viscosity vs. composition curves for blends of HDPE and LDPE at 190°C. o – experimental; —— calculated using eq. (2).

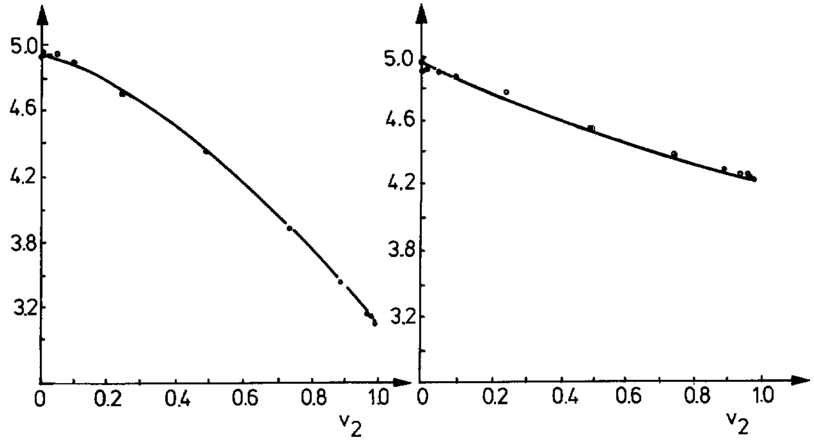

Figure 2. Viscosity vs. composition curves for blends of HDPE with LDPE (a) and of LDPE with LDPE (b). o – experimental ; —— calculated using eq. (2).

viscosity vs. shear stress equations for blend and pure components, v_1 and v_2 are the volume fractions of components, and $\mathcal{J}$ is a parameter. The value of parameter $\mathcal{J}$ may be calculated from the experimental flow curves for pure components and for the blend consisting of equal volume fractions of both components. This value allows then calculation of viscosity for blends

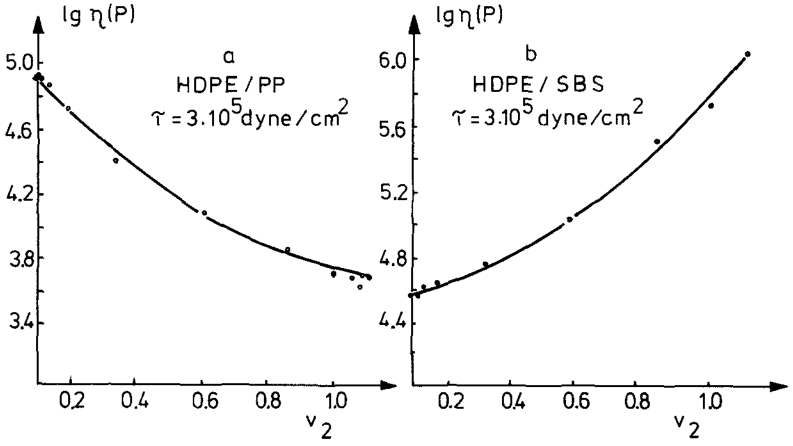

Figure 3. Viscosity vs. composition curves for blends of HDPE with PP (a) and of HDPE with SBS block copolymer (b). o – experimental ; —— calculated using eq. (2).

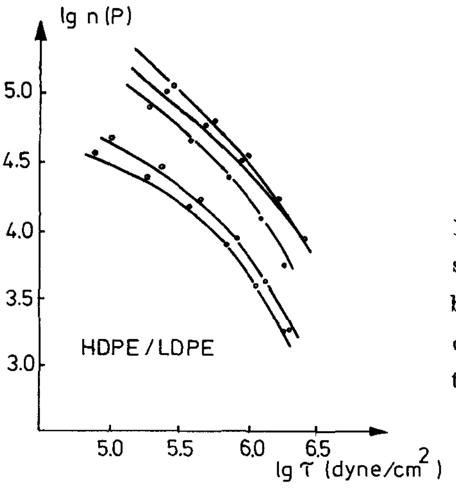

Figure 4. Viscosity vs. shear stress curves for different blends of HDPE and LDPE. o – experimental ; —— calculated using eq. (2).

of other compositions. Identical results are obtained using an equation of the form :

$$\lg\eta = v_1^2 \lg\eta_1 + 2\gamma v_1 v_2 (\lg\eta_1 \lg\eta_2)^{1/2} + v_2^2 \lg\eta_2 \qquad (2)$$

The utilization of this equation is much more correct because it corresponds to the requirement of correlating some rheological functions with

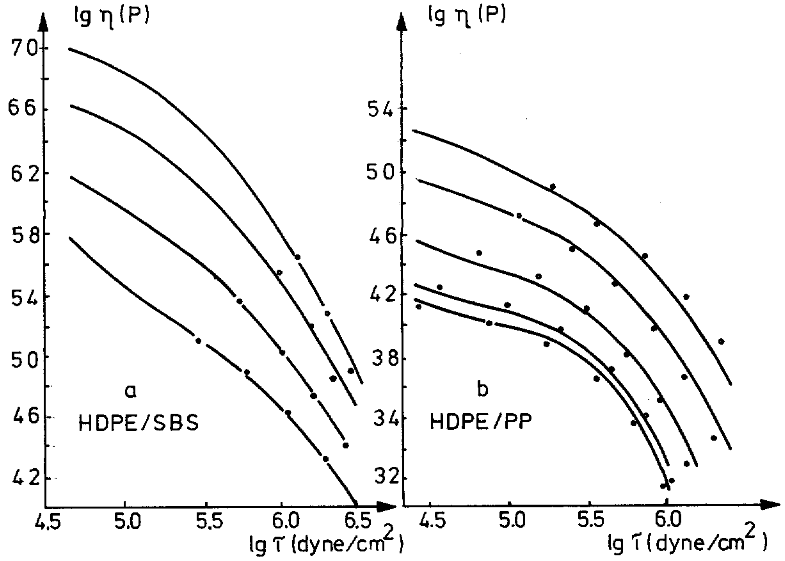

Figure 5. Viscosity vs. shear stress curves for different composition blends of HDPE with SBS block copolymer (a) and of HDPE with PP (b). o – experimental ; — calculated using eq.(2).

the square of component volume fractions. Besides, for the first model the parameter γ can be positive or negative and the sign change is not correlated with the nature of blend components. The second model ensures positive values of γ for all blends investigated. For HDPE / LDPE blends, the parameter γ in the second equation may be correlated with component viscosities using an equation of the form :

$$\gamma = 0.53613 + 0.72537\ R - 0.21682\ R^2 \qquad (3)$$

where

$$R = \lg \eta_{HDPE} / \lg \eta_{LDPE}$$

The agreement between calculated and experimental values is satisfactory. Larger errors are observed at the extremes of the composition range, due to the fact that the mathematical form of the model does not fit the sigmoidal shape of most of the viscosity – composition curves. Good results have been obtained for HDPE / HDPE, HDPE / LDPE, LDPE / LDPE, HDPE / PP, HDPE / SBS, PS / SBS blends, as illustrated in the Figures 1 – 5.

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