Flow Induced Morphology Development during Melt-Mixing in PP/EVA Blends

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CERTIFICATE

This is to certify that the thesis entitle 'Flow Induced Morphology Development during Melt-Mixing in PP/EVA Blends' being submitted by Mr. Sandeep Tyagi to the Indian Institute of Technology, Delhi for the award of degree of Doctor of Philosophy, is a record of bonafide research work carried by him. Mr. Sandeep Tyagi has worked under my guidance and supervision and has fulfilled the requirements for the submission of this thesis, which to my knowledge has reached the requisite standard. The results contained in this thesis have not been submitted, in part or full, to any other University or Institute for the award of any degree or diploma.

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Dedicated to

My Sister

Rashmi

&

My Daughter

Tisha

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ABSTRACT

The general aim of this study is to provide better understanding of the role of composition, component properties and processing conditions in morphology development during processing of polymer blends. This study reveals the development of morphology during blending of polymers in terms of domain size variation of the dispersed phase. Since the morphology influences the physical properties of polymer blends, it is important to be acquainted with the basic mechanisms of morphology evolution and the way it develops in the complex flow fields of industrially processing equipments. Though the mechanisms of droplet dispersion and coalescence are well known for Newtonian fluids, there is still a lack of fundamental understanding for the same in case of non-Newtonian polymers.

Polypropylene and ethylene vinyl acetate were used to carry out the melt-mixing studies in the extruder, in order to analyse the evolution of microstructure in the real processing operation. Processing of polymer blends was carried out to clearly understand the morphology evolution as affected by the rheological properties of the constituting components, blend compositions and processing variables from initial stage to the final stage in the extruder. Characterization of the dispersed phase (EVA) was carried out on the samples obtained from the screw channels after screw-pullout experiment. Different behaviour towards the polarised light (birefringence) of the PP and EVA phases was used to analyze the striations thickness distribution of the minor phase (EVA) in the feed zone of the

extruder whereas, droplet size distribution analysis in the metering zone was carried out using scanning electron microscopy by selectively etching out the EVA phase. For total understanding of the melt-mixing process, investigations were also made across the each channel sample by dividing the samples in terms of upper and lower layers along the channel depth.

Early stages of morphology development involve the formation of striations of the dispersed phase. Striation thickness measurements have been used in this study to express the state of mixing in terms of the dispersed phase domain size the distribution in the feed zone along the length of the extruder. The striations developed at the onset of melt-mixing have broader size distribution that is random in nature and skewed towards larger size range. As mixing proceeds, the decrease in striation size has been found to be associated with low values of variance, distribution being narrow and skewed more towards smaller size range. This decrease in the average striation size is associated with a regular decrease in standard deviation. Moreover, a critical size range has been identified as size larger than 100-125μm for stretching to be highly effective. Striations having thickness greater than this critical value show sharp decrease in their size, both in the upper and the lower layer. The percent of reduction of size also differs in each layer. It is interesting to note that, the rate of decrease in upper and lower layer become almost equal as the striation size goes below 100-125μm, interestingly. At the end of the feed zone, the striation reaches the critical size range of 20-40μm before the break-up process takes place.

The comparison of evolution of morphology during melt-mixing has been made for the different compositions of the blends. It has been found that, for all the three compositions i.e., 70/30, 80/20 and 90/10 wt.% PP/EVA, the average striation size decreases with decrease in concentration of dispersed phase as well as along the length of the extruder. The effect of viscosity ratio on the melt-mixing process in terms of striation size and striation size distribution has also been studied for blends with four selected viscosity ratios. Striation thickness does depend upon the viscosity ratio and showed sharper decrease along the length of the feed zone when the viscosities of both the phases were high (PP/EVA blend with viscosity ratio of 0.37). The effect of screw rotational speed on the morphology development indicates the presence of higher average striation size at low screw rpm as well as decrease of striations at a much faster rate than that obtained by processing at relatively higher rpm. A critical size range has been found to be present irrespective of blend composition, viscosity ratio and different screw rotational speed. The total process of morphology development seems to follow a typical behaviour that has been explained on the basis of "two-zone model" considering upper layer as strong zone and lower layer as weak zone.

In the metering and die zone of the extruder, there was reduction by one order of magnitude in the domain size and then it does not decrease much over the length of the metering zone. There is, however, shift in the distribution towards the narrow size range, indicating the homogenization process as a result of break-up and coalescence. In the die zone, oscillations in the droplet size was observed in some cases showing occurrence of emulsification process. Log normal model has been found to fit satisfactorily to the droplet distribution, both in the metering and die zone.

Finally the break-up process observed in the extruder environment as well as in the rheological experiments, has been analyzed and it was found that droplet size developed is in good agreement with the theory reported in literature. Palierne's model was found to describe the linear viscoelastic properties of the blends satisfactorily. The model of Peters, Hansen and Meijer (PHM) was used to predict the first normal stress difference of the blends under transient flow. Morphological evolution of the blends under stepwise increase in shear rate was obtained and good predictions with PHM model were observed.

Flow induced morphology development has been successfully correlated using log-normal distribution model and the models based on rheological responses. The present study is directly applicable for the development of tailor made morphology in polymer blends.

CONTENTS