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## ENCAPSULATION IN MULTIPHASE POLYMERS: ROLE OF VISCOELASTICITY, GEOMETRICAL AND INTERFACIAL PROPERTIES

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The aim of the present work has been to gain a fundamental understanding of the mechanisms governing encapsulation in the multiphase systems as a blend or multilayer structures. This phenomenon was recorded by optical observations as a function of the viscosity ratio, elasticity and the polymer/substrate interfacial tension. The study rendered it possible to decouple the influence of the viscoelastic parameters to shear, thereby highlighting a number of macroscopic effects that were governed by the interdiffusion or reaction of macromolecular chains at the interface:

- The investigation of model Newtonian polymers based on poly (dimethylsiloxane) of varying molar masses showed that the viscosity ratio of the material was a key parameter and that it had a considerable influence on the kinetics of encapsulation phenomenon. The encapsulation of two drops or molten particles placed in close vicinity of each other was monitored with a CCD camera at regular intervals time. An experimental illustration was given to confirm that the less viscous polymer tended to encapsulate its more viscous counterpart but at low shear rate. The effect of viscosity was more complex than initially supposed and appeared to be linked to the interfacial tension. The interfacial tension played a role for the more viscous materials at the same temperature and was found to accelerate or slow down the kinetics depending on the spreading parameters. On the other hand, this work showed that the viscosity ratio coupled to drop geometry of the material were a key parameter and that it had a considerable influence on the encapsulation kinetics. At a low shear rate, the small drop was encapsulated by the bigger one. When of equal geometry, a high surface contact area interfacial area/volume allowed the chains to diffuse, thus increasing the density across the interface/interphase. Meanwhile, these results also confirmed that the viscous dissipation mechanism (encapsulation at high shear rate) given in the literature was not the sole mechanism present to explain the phenomena.

- In a second step, the kinetics of encapsulation of the high molecular weight based on PVDF/PMMA were assessed at several temperatures and compared to the corresponding results for PVDF/PVDF and PMMA/PMMA used as references. The present asymmetric, viscoelastic system is completely compatible and fully characterized. Furthermore, no encapsulation phenomenon was seen despite the difference in elasticity and surface tension for PVDF/PMMA with the same viscosity as compared to the symmetrical references. The encapsulation appeared to be hindered by the interdiffusion process in the case of compatible pair system despite their elasticity and surface tension contrast. The observed development represented the signature of the kinetics of diffusion where the macromolecular chains intertwined and gradually became replaced by a more robust interphase. The present phenomenon has been confirmed by the rheological investigation of the present bilayer systems.

- Finally, the same approach was applied to functionalized polymers to investigate the effect the effect of physico-chemical affinity. Two pairs of asymmetrical reactive polymers were selected. They are based on PE-GMA (Glycidyl methacrylate)/PVDF-g-AM (Maleic anhydride) as well as a nonreactive material based on PE/PVDF. The observed phenomena and the results were analyzed based on the physicochemical mechanisms involved in the diffusion/reaction interfaces. The results of an optical investigation of the encapsulation kinetics of two drops corroborated the data obtained through rheology of the bilayer systems. Indeed, no encapsulation was noted in these cases despite that the zero shear viscosity ratio of PE-GMA/PVDF-g MA was close to 1.5. The triggered copolymer at the interface hinders the encapsulation kinetic. On the contrary, PE/PVDF with a higher viscosity ratio confirms that the PVDF tend to encapsulate the PE. The results obtained by an optical investigation of two drops corroborated rheological data of the bilayer systems. The observed phenomena and the results were analyzed based on the physicochemical mechanisms involved in the diffusion/reaction interfaces. This seemed to confirm



that the parameter should be coupled to the physico-chemical affinity and viscoelastic parameters in order to give a better understanding of encapsulation phenomenon in multiphase systems. Hence, for a reactive system, the encapsulation kinetic could be reduced or eliminated by the creation of a copolymer at the interface.