

Title	非相溶高分子間における低分子化合物の相間移動
Author(s)	NAWAPHORN, KUHAkongKIAT
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Description	Supervisor:山口 政之, マテリアルサイエンス研究科, 博士

## Interphase Transfer of Liquid Compound between Immiscible Rubber Pairs

Blending of at least two different polymers is a general method to provide required properties of a final product. In general, the addition of a low-molecular-weight compound as a third component is often carried out in industries to enhance the performance, such as curative agents, stabilizer, and plasticizer. The distribution of these third components in a blend has to be seriously considered because it decides the quality of a product. The miscibility mismatch between a third component with each polymer can sometimes lead to an uneven distribution. This phenomenon occurs with the migration of a third component from one polymer phase to another acrossing the boundary of phases, i.e., interphase transfer, which has been reported by several researchers. Nevertheless, less attention has been paid on the effect of the temperature on the transfer phenomenon.

Here, the temperature dependence of the distribution state of a third component in immiscible blends is focused, which has not been performed at the best of my knowledge. Since the amount of a liquid compound as a third component can change with ambient temperature, it is possible to control the glass transition temperature  $T_g$  of each phase in a blend.

The interphase transfer of di-2-ethylhexyl adipate (DOA) plasticizer between amorphous polyolefins of ethylene-propylene copolymer (EPR) and poly(isobutylene) (PIB) is reported using laminated sheets. The difference in the temperature dependence in the interaction parameter between DOA with each polymer is found to be a strong driving force for the transfer. The obtained results show that more DOA stayed in the EPR phase at low temperature, i.e., at  $-20\text{ }^{\circ}\text{C}$  of annealing. In other words, some amounts of DOA transfer from PIB phase to more favorable EPR phase when the annealing performs at  $-20\text{ }^{\circ}\text{C}$ . Since the DOA amount in each sheet is determined by the ambient temperature, the change of  $T_g$  in each sheet could also be changed. The peak temperature of the dynamic tensile loss modulus, defined as  $T_g$ , of EPR is found to be located at lower while PIB shows higher  $T_g$ , owing to the uneven distribution of DOA. Moreover, the opposite transfer direction is observed when the annealing treatment is performed at  $40\text{ }^{\circ}\text{C}$ ; some amounts of DOA migrate from EPR to PIB.

Besides the plasticizer, uneven distribution of a coumarone-indene copolymer with low-molecular-weight, which is in a liquid state at room temperature, is studied as a third component. In this study, natural rubber (NR) and PIB are selected for an immiscible pair. After separation, it is found that more tackifier locates in PIB at  $-20\text{ }^{\circ}\text{C}$  annealing, whereas the tackifier migrates to the NR phase at  $40\text{ }^{\circ}\text{C}$ . Because of the transfer, NR shows a lower  $T_g$  after annealing at  $-20\text{ }^{\circ}\text{C}$  and a higher  $T_g$  when annealing was performed at  $40\text{ }^{\circ}\text{C}$ . The endothermic peak from DSC measurement at around  $0\text{ }^{\circ}\text{C}$  demonstrates that the crystallization of NR is responsible for the transfer phenomenon at low temperature.

Considering that most tires are composed of at least two rubbers with one or more liquid compounds, the concept of this research can be applicable to produce "all-season tire".

Furthermore, the interphase transfer phenomenon is highlight to be applied for controlling the temperature dependence of the transparency, i.e., thermochromic property, of an immiscible polymer pair composed of ethylene-vinyl acetate copolymer (EVA) and poly(vinyl butyral) (PVB). It is indicated that the refractive index of each separated sheet is greatly affected by the ambient temperature owing to the DOA transfer phenomenon. At high temperature, the DOA migration from PVB to EVA is accelerated, in which the refractive index of EVA is lower than that of PIB. On the contrary, a large amount of DOA is preferably resided in PVB at low temperature.

**Keywords:** interphase transfer, immiscible polymer blend, uneven distribution, plasticizer, tackifier