Research Activity Report **2013.10.30**

Research title		**Crosslinking Effect in Amorphous Polymers on Heat Conduction Properties**
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Visiting institution		Rensselaer Polytechnic Institute, USA
Visiting period		April 2012 - March 2013
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	Title	Professor

Summary of Collaborative Research Activities

Crosslinking in polymer materials has been extensively studied in order to modify mechanical and chemical properties. Although abundant studies concerning the mechanical behavior of crosslinked polymers have been reported, there seems relatively little research on the effect of crosslinking on thermal transport properties. Therefore, in the present study, we are devoted to elucidating how a crosslink formation affects the thermal transport at the molecular level.

In order to gain a quantitative understanding of the relationship between crosslinking and thermal conduction, MD simulations on model amorphous polymers including polyethylene (PE) and atactic polystyrene (PS) were performed. Two types of PE models with different hydrocarbon chain lengths comprised of 50 and 250 carbons in a single chain (referred to as C50 and C250 hereafter) were adopted. A snapshot of an equilibrated configuration for the polystyrene system is shown in Fig. 1. In the nonequilibrium MD (NEMD) simulations, the constant heat flux was imposed (see also Fig. 1) and thermal conductivity can be monitored by picking up a temperature gradient in the MD system. When configuring crosslinked polymers, the crosslink bonds were formed only between the different polymer backbone carbons in both the PE and PS systems. The number of crosslink bonds introduced into the system was determined using a degree of crosslinking (DC).

As a result, thermal conductivity increases more or less linearly with crosslink concentration in the amorphous PE systems as shown in Fig. 2. On the other hand, in the PS case, the variation in thermal conductivity is insignificant for all the

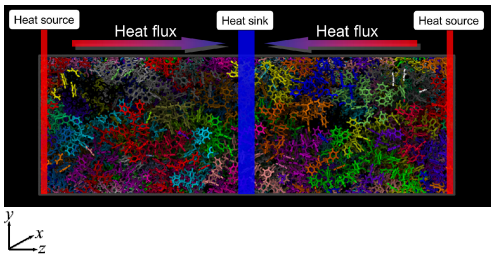

Fig. 1 : A snapshot of the polystyrene system. Each molecule (chain) is differently colored for clarity.

crosslink concentrations studied. This striking difference in thermal conductivity would be due to the highly heterogeneous PS structure including phenyl side groups providing the interaction and configuration heterogeneity into polymer network.

In order to elucidate the underlying mechanism for the effect of crosslinking, we decomposed macroscopic heat flow into microscopic "building blocks", i.e., energy transfer modes associated with various bonded and nonbonded interactions. From the result in the PE case, it is evidently observed that the change in the contribution of nonbonded interactions is more or less insignificant. On the other hand, the contribution of bonded interactions increases with increasing crosslink concentration. This indicates that introducing crosslink bonds opens new heat paths and directly contributes to the increase in thermal conductivity of crosslinked PE.

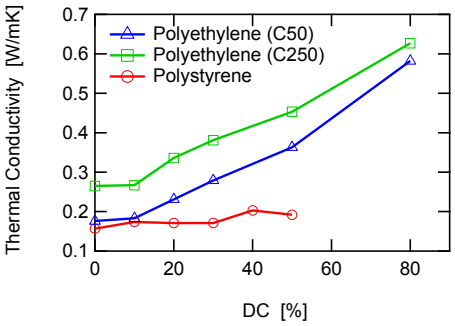

Fig. 2 : Thermal conductivities in the C50 PE (blue marked line), C250 PE (green marked line), PS (red marked line) as a function of a degree of crosslinking.

Publications

1) Journal or conference papers with full paper review (including review papers, invited papers, etc.)

[1] Gota Kikugawa, Tapan G. Desai, Pawel Keblinski, and Taku Ohara: Effect of crosslink formation on heat conduction in amorphous polymers, *J. Appl.Phys.*, Vol. 114, (2013), 034302.

[2] Gota Kikugawa, Pawel Keblinski, and Taku Ohara: Heat Conduction in Amorphous Polymers with Crosslink Formation, *Proc. 4th International Symposium on Micro and Nano Technology*, (2013), ID-71 (in CD-ROM).

2) Conference papers or presentation without full paper review

[3] Gota Kikugawa, Pawel Keblinski, and Taku Ohara: Thermal Energy Transfer in Amorphous Polyethylene with Cross-Link Formation, *2012 Rensselaer Nanotechnology Center Research Symposium*, (2012).

[4] Gota Kikugawa, Pawel Keblinski, and Taku Ohara: Heat Effect of Cross-linking on Heat Conduction of Amorphous Polymers, *2013 MRS Spring Meeting and Exhibit*, (2013).

3) Patent, award, press release etc.

Not applicable