学 位 論 文 の 要 約

博士の専攻分野の名称　博士（生命科学）or（薬科学）or（ソフトマター科学）or（臨床薬学）　氏 名 鄭 庸

学 位 論 文 題 名

Study on the Effect of Polymer Dynamics and Phase Separation on the Mechanical Performance of Double Network Materials
（ダブルネットワーク材料の力学性能に及ぼすポリマーダイナミクスと相分離の影響に関する研究）

The understanding towards the factors affecting and how they are affecting the mechanical performances of soft materials is essentially of great importance to the extensive applications of soft materials. In this doctoral dissertation, the author has systematically studied the effect of polymer dynamics and phase separation on the mechanical performances of double network (DN) materials systems.

The author first focused on the effect of polymer dynamics on the deformation and fracture behaviors of DN gels. To control the polymer dynamics, the author utilized the ethylene glycol (EG)/water mixtures and glycerol/water mixtures as solvents to systematically tune the solvent viscosity while keeping comparable solvent quality as like water to the constitutive polymers of DN gels. The author observed that the tensile behaviors of unnotched DN gels remained unchanged while the fracture energy measured by pure shear test of notched specimens decreased significantly with increasing solvent viscosity. This reduction in fracture toughness was found directly related to the decrease of the yielding zone size around the crack tip at increased solvent viscosity. These results suggested that viscous solvents control the dynamics of the stretchable network, which play an important role in the load-transfer process ahead of crack tips.

In addition to effect of polymer dynamics, the author also systematically investigated the phase separation structure in DN elastomers. To control the phase separation, the copolymer made from ionic monomers (*i.e.*, AMPS) and non-polar monomers was used as the first network and homopolymer made from non-polar monomers was used as the second. The phase separation structure in the DN elastomers was clarified by small-angle X-ray scattering (SAXS) and transmission electron microscopy (TEM). The author found that the nanophase-separated structure of DN elastomers can be controlled by varying ionic monomer fraction f_{AMPS} and crosslinking density (C) in the first copolymer network. In addition to controlling the f_{AMPS} and C, increasing network multiplicity can also tune the nanophase separation structure accordingly.

After characterizing the phase separation structure, the author then investigated the effect of the nanophase-separated structure on the mechanical performance of DN elastomers. The author systematically tuned the nanophase separation by changing the ionic-monomer fraction f_{AMPS} in the first network while maintaining almost the same primary double-network structure. The author investigated the effects of the nanophase-separated structure on the internal fracture and energy dissipation during tensile deformation in DN elastomers using tensile tests and cyclic tensile tests. The effect of the nanophase-separated structure on the fracture and fatigue resistance of the DN elastomers was also studied. The author elucidated that the interplay mechanism between the double network structure and the nanophase separation structure enhances the elastic modulus, strength, fracture toughness, and fatigue resistance all together. The interplay mechanism between the double network effect and nanophase-separation effect clarified in this dissertation showed that superimposing high-order structures in double network materials could bring further strengthening and toughening of the materials. Such strategy should be a universal approach for elastomers and gels in principle, which opens broad research opportunities, and will facilitate more researches as the next stage of the

strengthening and toughening of the soft materials.

All the phase-separated DN elastomers exhibit remarkable yielding and necking behaviors as typical, non-phase-separated DN hydrogels. For potential widespread applications of phase-separated DN elastomers, it is of great practical importance to understand yielding criteria of phase-separated DN materials and control their yielding point by their compositions. The author further investigated the yielding criteria of phase-separated DN materials. The author chose f_{AMPS} = 0.1, 0.5 and 1.0 as typical examples, and controlled the prestretch ratio and the phase separation of the first network while keeping other conditions constant by performing multiple swelling/polymerization steps to create samples with multiple networks (MN) from double to quintuple or sextuple. The yielding criteria clarified in this dissertation gave clear insight into the structure and yielding mechanisms of MN materials, which should be useful for designing tough materials with desired mechanical properties for various applications.

In conclusion, the author has extensively studied the effect of polymer dynamics and phase separation on the mechanical performance of DN materials.