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Why many polymers are so fragile: a new perspective VLADIMIR NOVIKOV, Univ. of Tennessee Knoxville, CECILE DALLE FERRIER, Laboratoire Leon Brillouin, ALEXANDER KISLIUK, ORNL, LIANG HONG, Shanghai Jiao Tong University, GIOVANNI CARINI JR, UOS di Messina, GIUSEPPE CARINI, GIOVANNA DANGELO, Universita di Messina, CHRISTIANE ALBA-SIMIONESCO, Laboratoire Leon Brillouin, ALEXEI SOKOLOV, Univ. of Tennessee Knoxville and ORNL — Many polymers exhibit much higher fragility than liquids of small molecules. Its mechanism remains a puzzle. We analyzed correlation of many properties of polystyrene to its fragility for samples with various molecular weights (MW). We demonstrate that these correlations work for short chains, but fail with increase in MW. Fragility of the viscosity that is determined by chain relaxation follows the correlations at all molecular weights. These results suggest that the molecular level relaxation follows the behavior usual for small molecules even in polymers, while segmental relaxation has unusually high fragility. We speculate that many polymers cannot reach an ergodic state on the time scale of segmental dynamics due to chain connectivity and rigidity. This leads to decrease in accessible configurational entropy upon cooling and results in higher fragility of segmental relaxation. This scenario provides a new insight in polymer dynamics: the role of ergodicity time and length scale. [1] C. Dalle Ferrier, et al, J. Chem. Phys. 145, 154901 (2016).

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