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Entropically Driven Exfoliation of Clay in a Clay/Polymer Nanocomposite HORST WINTER, KATIE LANIA, FEI LI, XIAOLIANG WANG — Dynamic mechanical spectroscopy was used to monitor the maturing of structure in a clay/polymer nanocomposite. The rheological observations show that the classical mechanism of exfoliation by diffusion has to be modified. A new exfoliation mechanism is proposed as follows: when end-functionalized ("sticky") macromolecules anchor at the surface of clay particles, the thermal motion of the polymer is restricted due to the presence of the solid surface. This gives rise to an entropic pulling force on the clay surface which peels clay sheets away from the clay particles (stacks of clay sheets). The clay, organically modified with macro-counterions, is only weakly connected internally so that the macromolecular entropic outer forces can overcome the internal cohesion. The entropic force increases with temperature due to the increased thermal motion, but then decreases again when macromolecules begin to detach. Polymer molecules and clay connect into a sample-spanning network with increasing modulus and decreasing relaxation time as the clay exfoliation proceeds and more and more clay surface becomes accessible to the polymer.

Horst Winter

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