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A new molecular theory beyond tube model to describe cohesive breakdown in nonlinear flow of entangled polymers. SHI-QING WANG, University of Akron — When an entangled polymer is subjected to shear or extensional flow at a rate of deformation (RD) much greater than its dominant relaxation rate (dRR), it may not flow homogenously all the way to the limiting strain of (RD/dRR)before it suffers cohesive failure. What keeps the chains entangled is an essential question to answer before an appropriate theory of polymer flow can be established. Unlike the tube model that assumes presence of an infinitely high energy barrier preventing escape of chain entanglement, our theory [1] recognizes a finite barrier height given by kT(M/Me) for a polymer whose number of entanglements per chain is (M/Me). A second essential ingredient is to realize that a sufficiently high level of elastic force can be generated per chain by the externally imposed flow. This elastic force can overcome the entanglement (cohesive) force as a rate-activation process, leading to the onset condition for the cohesive breakup either during flow or upon cessation of flow. Flow produces frictional inter-chain interactions among all entangling chains. These interactions also resist constitutive disintegration, delaying the onset of cohesive collapse to a larger strain. A higher level of cohesive strength results from the very flow deformation that could eventually produce enough internal (elastic) forces to destroy the cohesive structure made of chain entanglement. [1] Phys. Rev. Lett. 97, 187801 (2006).

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