

MAR15-2014-020252

Abstract for an Invited Paper
for the MAR15 Meeting of
the American Physical Society

New insights on cellulosic ether hydrogels

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Aqueous hydroxypropylmethylcellulose materials (HPMC) often have much lower hot gel moduli (<10 Pa) relative to those (3,000 Pa) of aqueous methylcellulose materials (MC) at end-use conditions (<2 wt.%, 90 °C), and these lower moduli limit their use in applications. The origin of their lower moduli is suspected to arise from the order of two thermal transitions when warming. One transition, thought to involve a chain conformation transition, is referred to here as chain collapse. Another, thought to involve the self-assembly of chains into a three-dimensional physical network, is referred to as gelation. Often, chain collapse is thought to proceed gelation when slowly warming aqueous commercial HPMC materials from 5 to 90 °C at 1 °C/min, while the opposite order is thought to occur for many aqueous commercial MC materials. Chain collapse is identified as a sharp drop in the solution viscosity at pre-gel temperatures as T rises. The insensitivity of the chain collapse temperature to HPMC concentration is used to argue that this thermal event is distinct from gelation. These concepts are supported with the preparation of two developmental HPMC materials with similar MW and substitution levels (DS and MS). One HPMC material, prepared by a unique process, is designed to reverse order of the thermal transitions. This HPMC material is found to exhibit high hot gel moduli similar to those of aqueous MC materials; moreover, its gel is able to form synerese fluid as it contracts in size when warmed. The gel contraction is thought to be a manifestation of chain collapse.