Comprehensive Modelling of the Experimental Temperature and Stress Response of Time-Dependent Materials

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Summary: The stress- and temperature-dependent non-linear creep behaviour of polymers and other materials is quantitatively modelled as a superposition of a wide range of activated motions at the molecular scale, covering wide but well-defined space and time scales. The resulting time, temperature and stress dependences are coupled (not fully separable) except at low stress values, where linear viscoelastic behaviour is directly obtained, improving on previous (approximate or questionable) time-temperature and time-stress superposition or equivalence relationships. The behaviour has been shown to have a strong cooperative nature, which may be interpreted in terms of varying clusters of identifiable structural elements within the material. Reasonably good descriptions of the experimental creep behaviour of both amorphous and semicrystalline polymers have already been achieved, but this work concentrates on the behaviour of an amorphous polycarbonate (PC), including the physical characterization and modelling of its retardation time spectrum.

Keywords: cooperative effects; creep; macromolecular dynamics; modelling; polycarbonates

Introduction

The comprehensive and accurate modelling of the (thermal, viscoelastic, etc.) behaviour of time-dependent (polymer and other) materials still remains a challenge to materials science. Non-linear creep behaviour of amorphous and semi-crystalline polymers is just one aspect of such challenge, on which this contribution is focused.

Previous and current interpretations and modelling of the creep behaviour of polymers have relied and still rely mostly on empirical or semi-empirical formulations, without directly taking into account the physical (molecular) underlying mechanisms, namely the conformational and other transitions, at the molecular scale, responsible for the materials' non-linear viscoelastic behaviour.[1–6] A feature of most of these descriptions is the separation of the effects of stress and time, in addition to that of temperature, into separate factors making up the strain, $\varepsilon(t,\sigma_0,T)$, or creep compliance, $D(t,\sigma_0,T) = \varepsilon(t,\sigma_0,T)/\sigma_0$, which is only an approximation. The emphasis has been on time-temperature and time-stress superposition or equivalence relationships, for interpolation and extrapolation purposes, which may be valid only within limited operating ranges of time, temperature and stress, at best. The approach adopted in this work will instead aim at a predictive model of the behaviour.

Dynamic Modelling

References 7–10 developed the detailed dynamics of gauche-trans conformational

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