Time-temperature and time-stress correspondence in non-linear creep. Experimental behaviour of amorphous polymers and quantitative modelling approaches

José R. S. André[1] *, José J. C. Cruz Pinto[2]

[1] Department of Mechanical Engineering, Technology and Management School, Guarda Polytechnic Institute, Avª Dr. Francisco Sá Carneiro, nº 50, 6300-559 Guarda, Portugal; http://www.ipg.pt; Fax 271 220 123; jandre@ipg.pt

[2] CICECO, Composite and Ceramic Materials Associate Laboratory / Department of Chemistry, University of Aveiro, Campus of Santiago, 3810-193 Aveiro, Portugal; http://www.dq.ua.pt; Fax 234 370 084; CPinto@dq.ua.pt

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Abstract: Non-linear creep is described by a non-simulative, analytical, dynamic molecular modelling approach. Elementary, molecular-scale, process-relevant frequencies are derived by adequate kinetic formulation. They follow almost exactly an Arrhenius-like behaviour with a range of activation enthalpies. Their relative contribution to the overall macroscopic behaviour of the materials is quantified to account for the materials' retardation time spectra and final non-Arrhenius behaviour. A new creep compliance equation is derived, yielding a fully coupled time-temperature-stress formulation, with long-term predictive capability. Experimental data for poly(methyl methacrylate) are analysed to identify the extent to which time-temperature and time-stress correspondence relationships may be valid, and it is shown that they are approximations (especially the latter), limited to narrow ranges of experimental variables, in contrast to the proposed model, which more reasonably fits the experimental behaviour.

1. Introduction and relation to previous work

Previous models for the interpretation of the creep behaviour of polymers are mainly empirical or semi-empirical, and do not directly take into account the physical (molecular) underlying mechanisms, namely the conformational and other transitions responsible for the material's non-linear viscoelastic behaviour. In fact, even the recent work by Kromm et al. [1] still describes the creep behaviour of a semi-crystalline polymer (ultra-high molecular weight polyethylene) by means of a Nutting-type equation, modified by a single Arrhenius-type exponential to account for the effect of temperature. Also recently, Sen et al. [2] presented fits to experimental creep data of purely phenomenological models combining Voigt-Kelvin units with Trantina's Nutting-type empirical model [3]. Various other contributions had a similar