Journal of Macromolecular Science, Part B
Physics

ISSN: 0022-2348 (Print) 1525-609X (Online) Journal homepage: http://www.tandfonline.com/loi/lmsb20

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To cite this article: V. I. Kartsovnik, R. Worlitsch, F. Hermann & Y. Tsoglin (2016) Calculation of the Viscosity of Polymer Melts Based on Measurements of the Recovered Rubber-Like Deformation, Journal of Macromolecular Science, Part B, 55:2, 149-157, DOI: 10.1080/00222348.2015.1119340

To link to this article: http://dx.doi.org/10.1080/00222348.2015.1119340

Accepted author version posted online: 20 Nov 2015.

JOURNAL OF MACROMOLECULAR SCIENCE®, PART B: PHYSICS
2016, VOL. 55, NO. 2, 149–157
http://dx.doi.org/10.1080/00222348.2015.1119340

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Calculation of the Viscosity of Polymer Melts Based on Measurements of the Recovered Rubber-Like Deformation

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ABSTRACT

On the basis of a model of polymer flow, considering the forces of entropic elasticity of extended macromolecules within the Eyring's concept, the relationships between the shear rate, shear stress, viscosity, and recovered rubber-like deformation were derived. The reduction of activation energy of the flow, by an amount proportional to the recovered rubber-like deformation, leads to an exponential decrease of viscosity with increasing shear rates; this nonlinear dependence of viscosity on shear rate (and shear stress) is defined as the viscosity anomaly of polymers. The measurement of deformation recovery after the cessation of polymer flow in the mode of constant shear rate or shear stress on a rotational viscometer confirmed the validity of the theoretical dependences.

ARTICLE HISTORY
Received 23 April 2014
Accepted 3 November 2015

KEYWORDS
activation energy; entropic elasticity of macromolecules; eyring's theory of viscous flow; recovered rubber-like deformation; viscosity anomaly

1. Introduction

A nonlinear dependence of viscosity on the shear rate and shear stress is observed for most polymer media–polymer melts and solutions and is called the anomaly of viscosity.[1] It is expressed in the decrease of the apparent viscosity, η, defined as the ratio of experimentally measured shear stress, τ, to the shear rate, $\dot{\gamma}$:`

$$\eta = \tau/\dot{\gamma}. \tag{1}$$

According to the theory of thermally activated flow of liquids of Frenkel,[2] an elementary act of the fluid flow process is the jumping of liquid molecules, as a result of thermal energy fluctuations, from one equilibrium position into another, neighboring vacant place—"hole," by overcoming the potential energy barrier. A certain activation energy, E_0, is required for this jump. In accordance with the theory of viscous fluid flow of Eyring,[3,4] the external force, τ, per 1 cm^2, that provides the fluid flow, affects the kinetic unit (atom, molecule, molecular segment of polymer chain) which is jumping into the vacancy—"hole," during the flow of the material. This decreases the activation energy by a magnitude proportional to the applied force, τ. Based on the theory of rate processes[4] concerning the non-Newtonian

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