YU ISSN 0015—3206
FZKAAA 12 (1) 55 (1980)

VISCOSITY OF LIQUID SELENIUM

A. H. ABOU EL ELA, H. H. A. LABIB* and K. A. SHARAF

Physics Department, Islamic Girls College, Nasr City, Cairo and Physics Department, Faculty of Education, Ain Shams University, Cairo, Egypt

Received 10 July 1979

Revised manuscript received 5 January 1980

UDC 532.13

Original scientific paper

The viscosity of selenium has been determined in the liquid state, where the rotating cylinder method was used. Experimental data show that the apparent activation energy of the viscous flow decreases with increasing the temperature. The value is 0.5 eV at the melting point and 0.26 eV at 300°C.

1. Introduction

The viscous flow behaviour of selenium has been the subject of several previous investigations. In some cases[1-3], these cover the high temperature range above the melting point, while in another[4], the flow behaviour in the vicinity of the glass transition point was explored, where Cukierman[5] had observed departures from newtonian flow in amorphous selenium. Moreover, experimental data had shown that the viscosity of liquid selenium is sensitive to small amounts of monovalent impurities. It is lowered by the addition of small amounts of chlorine, bromine, iodine and thallium. Since the high viscosity of selenium is attributed to the long selenium chains, it appears that the average chain length is reduced by these impurities. Furthre, the viscous flow was found to be very sensitive to change in structure.

The aim of the present work is to study the viscosity of liquid selenium at high temperatures above the melting point in order to clarify possible changes in structure, since in liquid selenium a chain structure seems to be preserved with a strong temperature dependence. The method used in the present work is the rotating cylinder method which is one of the best methods for measuring the viscosity, specially for a wide range of temperature, because of its high accuracy.

2. *Experimental*

Viscosity measurements were carried out using the rotating cylinder method[6], in which the damped oscillations of a cylinder filled with the liquid sample and suspended by an elastic filament were recorded. The viscosity was calculated by the method of successive approximations using Shvidkovskii equation[7]

$$\eta = \frac{0.8225\, M R^2 R^2 \sigma}{2\tau \left[(I + M R^2/2)\, \delta - I \frac{\tau}{\tau_0} \delta_0 \right]} \cdot d$$

where M is the mass of the liquid, I is the moment of inertia of the empty cylinder, R is the radius of the cylinder, δ and δ_0 are the logarithmic decrements of the damped oscillations of the filled and empty cylinder, τ and τ_0 are the oscillation periods of the filled and empty cylinder, and d is the density.

Density measurements were carried out using the thermometer method[6]; a certain mass of the liquid sample was placed in a specially shaped silica ampoule whose middle part is capillary with a scale allowing for measurements of volume at different temperatures.

3. *Results and discussion*

The temperature dependence of the viscosity of liquid selenium is shown in Fig. 1. It appears that the slope of the curve which corresponds to the apparent activation energy of the viscous flow decreases with rising temperature, the value is about 0.5 eV $\pm$ 00.5 eV at the melting point and 0.26 eV $\pm$ 0.05 eV above 300 °C. Fig. 2 shows the temperature dependence of the density of pure selenium, the density values agree very well with those of Glazov[6] and Ruska[8]. The derease of density with temperature may be attributed to a change in structure superimposed to the thermal expansion.

Bueche[9] has found, on the assumption of freely orienting chains in polymers, that the viscosity is proportional to the average chain length Z and the segmental friction factor F (the force required to pull a single segment through its surroundings at unit speed),

$$\eta = Z F \qquad (1)$$

For the free volume formalism[10], the segmental friction factor is

$$F = A\, T^{1/2} \exp\left[\frac{\gamma\, v^*}{\alpha\, \overline{v}_m (T - T_0)}\right] \tag{2}$$

where A is a constant, γ is a numerical constant accounting for the overlap of free volume ($\gamma \simeq 0.5$), v^* is the critical volume which must be available to a molecule before a diffusive motion, α is the coefficient of thermal expansion, $\overline{v}_m$ is the mean molecular volume, and T_0 is the temperature at which the free volume disappears (for melt polymers T_0 is close to the glass transition temperature T_g).

In liquid selenium the polymerization equilibrium is strongly depending on temperature, and as long as the average number of atoms in a chain is large, Z decreases with increasing temperature as

$$Z = Z_0 \exp(E/kT) \tag{3}$$

where Z_0 is a constant.

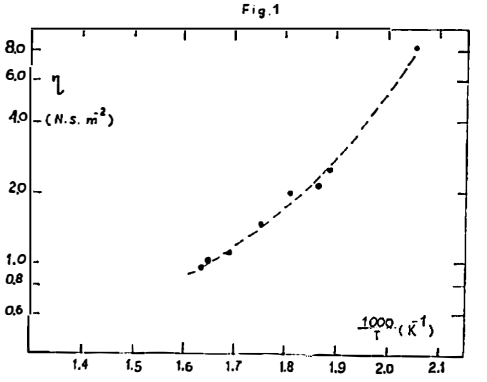

Fig. 1 Temperature dependence of the viscosity of liquid selenium.

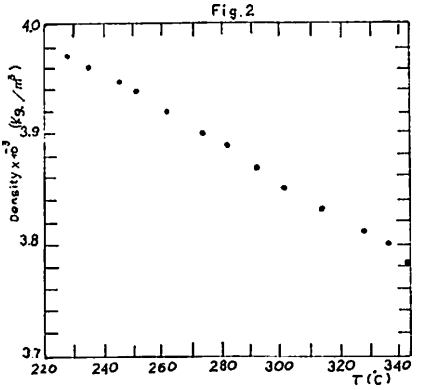

Fig. 2 Temperature dependence of the density of liquid selenium.

The valuc of the activation energy calculated from Fig. 1 (E = 0.5 eV) agree with the calculations of Keezer and Bailey[3] who had shown that the viscosity--chain length relation is described by the relation

$$\eta = 1.57 \cdot 10^{-7} Z^{1.45} \tag{4}$$

from which the chain length Z can be calculated. Fig. 3 shows the temperature dependence of the chain length for liquid selenium, the chain length decreases with the temperature.

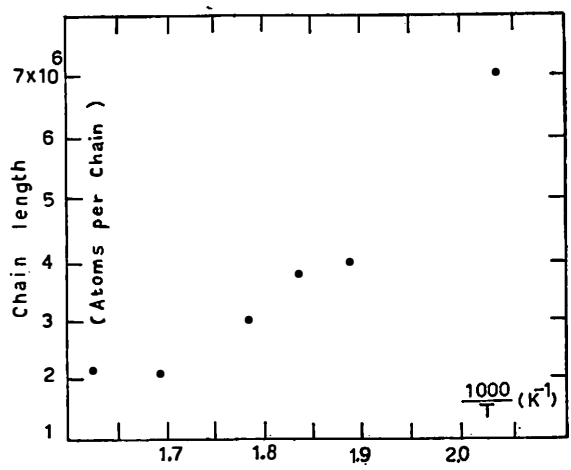

Fig. 3 The dependence of the chain length of selenium molecule on the temperature.

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VISKOZNOST TEKUĆEG SELENA

A. H. ABOU EL ELA, H. H. A. LABIB* i K. A. SHARAF

Physics Department, Islamic Girls College, Nasr City, Cairo and Physics Department, Faculty of Education, Ain Shams University, Cairo, Egypt*

UDK 532.13

Originalni znanstveni rad

Primjenom metode rotirajućih cilindara određena je viskoznost selena u tekućem stanju. Eksperimentalni podaci pokazuju da energija aktivacije viskoznog toka opada s porastom temperature. Njezina vrijednost je 0.5 eV na temperaturi taljenja, a 0.26 eV na 300°C.