

Yielding of Crystalline Polymers

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NOTE

Yielding of Crystalline Polymers

By Kazunori KITAJIMA

Compared with those of metals, yielding of crystalline polymers are characterized by large specific yield stress and activation energy which seem to be closely connected with the characteristics of crystal structure of polymers. In this paper an attempt is presented which explain these interrelations in the words of dislocation theory.

1) Experimental evidences

a) Evidences of crystalline plasticity

Frank and Keller (1958)⁽¹⁾ had deduced the operation of (100) [010] slip and (110) [$\bar{1}\bar{1}$ 0] twin systems in the deformation of pressed polyethylene, and Zaukelies (1962)⁽²⁾ presented an evidence of (010) [1 3 14] slip in the deformation of oriented Nylon. On the other hand, Takayanagi (1962 unpublished work) found that b-axis oriented specimen of polyethylen show more large residual strain than those of unoriented specimens after extension and unloading in the range of stress lower than the yielding (necking) stress.

b) Characteristics of stress-strain curves

Kobayashi (1962)⁽³⁾ and Takayanagi (unpublished work) reported the tensile tests performed on the oriented polyethylen in the range of various temperatures and strain rates. Using these data yield (necking) stresses are analysed by means of the stress dependent rate process equation

$$\dot{\epsilon} = Z \exp(-U/kT), \quad U = U_0 - v\sigma = U_0(1 - \sigma/\sigma_0),$$

and it is found that the yield stress extrapolated to 0°K σ_0 is about 10 kg/mm², and activation energy of yielding U_0 about 1eV. Taking elastic modulus $E \approx 100$ kg/mm², and lattice constant $b \approx 5 \text{ \AA}$, we find the specific yield stress $\sigma_0/E \approx 1/10$ and specific activation energy $U/Eb^3 \approx 1$ are very large compared with those of metals (Table I).

Table I

Kobayashi (1962)

$$U_0 = 2.06 \text{ eV}$$

$$\sigma_0 = 12.6 \text{ kg/mm}^2$$

Takayanagi

$$U_0 = 1.03 \text{ eV}$$

$$\sigma_0 = 20 \text{ kg/mm}^2$$

$$\frac{\sigma_0}{E} = 1 \sim 2/10$$

$$U_0/Eb^3 = 1.4 \sim 2.8$$

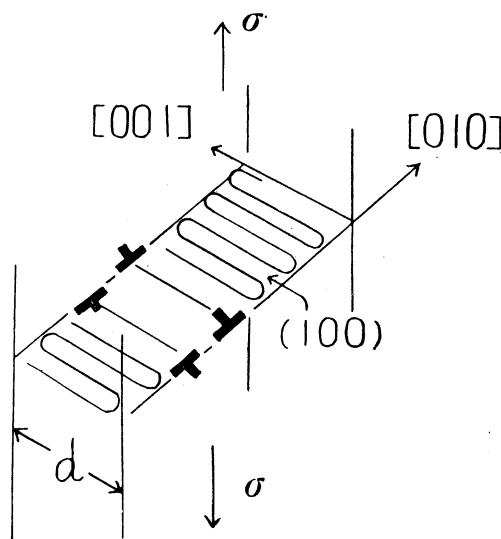


Fig. I. nucleation of dislocation pair.

2) Discussions

The experimental evidences a) mentioned above shows that deformation of crystalline polymers can also be interpreted by crystal plasticity essentially similar to metals. b) shows, however, detailed dislocation mechanisms are very different from those of metals. Zaukelies⁽²⁾ suggested that the mechanisms must be explained based on the crystal structure containing much defects such as point defects, dislocations and crystal boundaries, but no definite model mechanisms were proposed.

Then taking into account the characteristic "lamella" structure of crystalline polymers in which molecular chains are considered to be folded along c-axis into the thickness of about 100 Å, we now examine for instance (100) [010] slip as the representative mechanism in such crystals.

a) Judging from the nature of Van der Waals type binding forces, at first the magnitude of the Peierls stress of the dislocation is considered to be smaller than the order of $10^{-5} \sim 10^{-4}E$, and then the resistance for the motion of dislocation due to the interactions with other dislocations or point defects not exceed the order of $10^{-4} \sim 10^{-2}E$, and the activation energy of the movement be about $U/Eb^3 \leq 1/20$ as expected from the data of metals. From these facts we may deduce that the resistance for movement of pre-existing dislocations is not primary cause of yield stress of polymers. Moreover it is very difficult to conceive the operation of dislocation multiplication mechanism of Frank-Read or Orowan-Gilman type in the lamella structure.

b) It is instead considered to be rather preferable that the nucleation of dislocation pair in otherwise perfect lattice as observed in metal whiskers is the rate controlling mechanism particularly of yielding stress (see Fig. 1). In this model, however, the state of stress and displacement is assumed to be two dimensional because of the large anisotropy of the crystal structure, and the critical stress of nucleation is calculated to be $\sigma_0 \approx E/10$ and activation energy $U \approx E/4\pi \cdot b^2 d \log \left(\frac{\sigma_0}{\sigma} \right)^{(4)(5)}$, where $d \approx 20b$ is the thickness of the lamella. As easily proved this model is able to account the experimental values. Then the processes of deformation in the present view may be pictured in such a way, pre-existing dislocations firstly move and then some dislocations are nucleated at stress concentrated regins but the movement of dislocations once produced is not primarily responsible for the resistance to deformation. While at the stresses near the yielding stress dislocations are nucleated rather homogeneously, thus unfolding and necking result.

Operative system of nucleation must be determined by the critical stress and activation energy of the system. Twinning system (110) [110] is more probable one since magnitude of Burgers Vector b is as small as 1 Å.

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