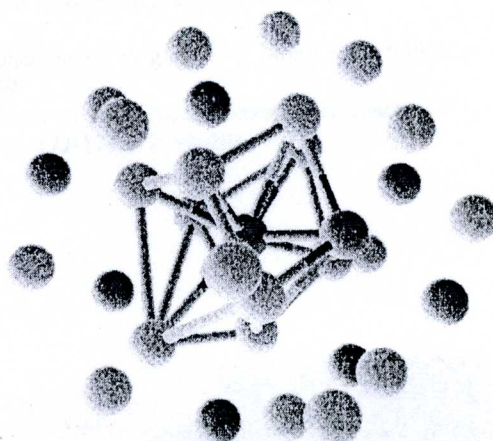


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## Sigmoid tendencies in the kinetics of natural physical ageing in $\text{As}_{10}\text{Se}_{90}$ glass

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It is shown that kinetics of enthalpy losses caused by prolonged storage of glassy  $\text{g-As}_{10}\text{Se}_{90}$  in a dark at room temperature during more than two decades (natural physical ageing) exhibits a well-expressed sigmoid behavior – the plateaus and steep regions. The microstructure mechanism of ageing explains well the observed stretch-exponential behavior in relaxation kinetics in terms of hierarchically-constrained mixed serial-parallel relaxation events having different atomic precursors. This kinetics can be decomposed into elementary components, each of them being single exponential.

The model of natural physical ageing in  $\text{g-As}_{10}\text{Se}_{90}$  is developed at the basis of detailed kinetics study. Structural data for Se-rich As-Se glasses suggest that physical ageing initiated by the elementary twisting of inner Se atoms within specific double-well potentials associated with high flexibility of Se bonds [1]. In the first approximation, these twisted Se atoms can be accepted as elementary Johari-Goldstein (JG)  $\beta$ -relaxation precursors. They should result in the appearance of aligned Se-based regions in a glassy network and, consequently, local free volumes released by twisted Se atoms. Appearance of such free volumes around collapsed Se-based regions is assumed to be accompanied by elastic strains in their immediate atomic surrounding. This initial stage is responsible for the lowering of glass transition temperature during first period of ageing. If the accumulated elastic energy is high enough, the further cooperative rearrangements occur, eliminating redundant free volumes from the considered local atomic regions, which lead to the lowering internal energy of the system at this particular scale. Happening at a larger scale, these relaxation processes should lead to general shrinkage of glassy network. Therefore, in subsequent stages of ageing, a general shrinkage of network takes place, which can be attributed to JG  $\beta$ -relaxation facilitated further  $\alpha$ -relaxation. This process is a reason for increase in the glass transition temperature and enthalpy losses in conventional differential scanning calorimetry measurements [2].

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2. R.Golovchak, O.Shpotyuk, A.Kozdras, *Phys. Lett.* **A370**, 504-508 (2007).