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VISCOELASTIC AND PLASTIC BEHAVIOUR OF METALLIC AND OTHER GLASSES NEAR THE GLASS TRANSITION

J. Perez, J.Y. Cavaille, S. Etienne and F. Fouquet

Abstract.- Results about micromechanical properties (internal friction and creep) of several glasses (metallic, semi-conductor and oxydes glass) are presented: they seem more characteristic of the vitreous state itself than the chemical nature of the glass. The assumption about the formation of shear microdomains leads to the interpretation of the results which might be a contribution to a better knowledge of the "homogeneous" deformation of vitreous solids.

I - INTRODUCTION

In the large deformation range, it is well known that two modes of deformation can be observed in glassy materials, particularly in metallic glasses: an homogeneous mode at high temperature and inhomogeneous slip at low temperature. The present paper is concerned by the former, studied in the low deformation range by measurements of micromechanical properties (internal friction $\tan \delta$, creep, stress relaxation...). Indeed, such measurements lead to informations about the mobility of structural units when a stress is applied onto a vitreous solid.

In these conditions, two relaxation phenomena are generally observed [1]: one ($\alpha$ relaxation) corresponds to the movement of nearly all the structural units leading to permanent strain (viscoplasticity); the other ($\beta$ relaxation) could be the consequence of limited (localised) movements of some of the structural units leading to recoverable strain (viscoelasticity).

In the first part, some results about both relaxation phenomena observed in the case of different vitreous solids will be shown; in the second part, an interpretation of such a behaviour will be proposed.

II - EXPERIMENTAL RESULTS

From three exemples (oxydes, semi-conductor and metallic glasses), we intend to show how such materials can present a behaviour more characteristic of the vitreous state itself than of their precise chemical nature.

II-1 - Oxydes glass

The viscoelastic properties of an oxydes glass have been measured between 200 and 550°C [2]; the glass transition observed by a dilatometric test is about 385 - 395°C.

The curves $\tan \delta$ in function of the temperature show in the whole frequency range (between $10^{-3}$ to $10^{3}$ Hz) a monotonic increase.

Figure 1
Relation between internal friction ($\tan \delta$) and frequency for different temperatures; curves: experimental results and open triangles: calculated values obtained from simplified relation (3): $J_R^s = 1$, $\omega_1 > 1$ and $\tau_1 = \tau_2$ [2].
A noticeable feature is shown in the figure 1 with the variation of $tg\phi$ versus frequency for several temperatures: for comparison, a straight line of slope 1 corresponding to the MAXWELLIAN viscous flow, is added: it appears that the actual behaviour of the material is the more different from the MAXWELL model, the temperature is low and (or) the period is small: this means that there is not only a simple viscous flow but also a viscoelastic deformation as it has been shown previously: indeed, a recoverable strain does exist when the load is removed during a creep test [3,4].

II-2- Semi conductor glass

Internal friction of metallic glasses is not yet well known: at temperatures lower than $T_g$, relaxation peaks are observed in the case of different alloys (see [7] for instance). In the glass transition temperature range a monotonic increase of internal friction is observed till the devitrification occurs, inducing a decrease of internal friction (see [8] for instance). Thus, the alloy Fe-P-C has been studied by YOSHIDA et al [9]: values of activation energy are obtained either directly from the curves $tg\phi(T)$ ($E_f$) or from the shift of these curves when the frequency is modified ($E_i$): $E_f$ is always higher than $E_i$ and this feature is generally explained in terms of distribution of relaxation times [8,9].

As with other glasses the viscoelastic behaviour is associated with viscous flow [10]: in the case of an alloy Fe-P-C(*) we have observed that both aspects are present at temperature high enough (figure 4). The transient part of the creep curves has been more particularly experimentally studied: the main following features can be put forward:

1) at all temperatures, the strain rate at $t = 0$ varies linearly with stress till $1,2 \times 10^9$ Pa; after the transient and chiefly at higher temperature.

(*) This alloy has been elaborated by the "laboratoire central de SAINT GOBAIN-AUBERVILLIERS".
the strain rate is not any more linear with stress in the same range.

\[ \dot{\varepsilon} = a \dot{\varepsilon} + b \sigma + c \sigma^2 \]

iii) As with macromolecular materials, Kohlrausch’s experiment can be made: this experiment is schematically shown in the figure 6 [11].

\[ \frac{\dot{\varepsilon}}{\sigma} = k \text{e}^{\frac{-E}{RT}} \]

To get a general understanding of the preceding results, the physical process by which the structural units move in stressed vitreous solid in the transition range, ought to be precised. In a previous work [12], an hypothesis was made: due to the external stress, local shear transformation are nucleated through cooperative movements of a group of structural units: in other words, the strain is the consequence of the transition from one configuration state to another in a microdomain (hereafter called shear microdomain or smd) whose size is temperature dependant: this size increases as the temperature is decreased as long as the metastable equilibrium state is obtained [12]. Such a transition leads to an increase of the enthalpy of the solid either because of the dilation accompanying the formation of the smd or due to the distortion of bonds at the frontier of the smd: in all cases this enthalpy increase gives a viscoelastic character to the strain. On another hand, this strain is limited by the growing of an internal stress \( \sigma_1 \) (dilation effect, bond distortions) proportionnal to the number of smd, i.e., the strain: of course \( \sigma_1 \) can decrease through structural relaxation: thus, the behaviour of the glass can be described by the relations:

\[ \dot{\varepsilon} = C_1 (\sigma - \sigma_1) \]  
\[ \dot{\varepsilon} = C_2 \dot{\varepsilon} - C_3 f(\sigma_1) \]
with $\varepsilon$ : non elastic strain ; $\sigma$: applied stress ; $C_1$, $C_2$ and $C_3$ constants. The function $f(\sigma_i)$ is related to the conditions of decrease of $\sigma_i$ by structural relaxation and may be complicated [11] : only the limit case of linear viscoelasticity ($f(\sigma_i)$ is reduced to $\sigma_i$) will be considered here. Hence, equations (1) and (2) correspond to the rheological model of BURGERS. In other respects, the relation between $\tan \phi$ and $\omega$, and KOLRAUSCH's experiment lead us to take into account of a distribution of relaxation time which must be the less wide the temperature is high: all the preceding considerations are taken into account by using the DAVIDSON and COLE relation giving the complex compliance:

$$J(\omega) = J_I \left[ 1 + (i \omega \tau_2)^{-1} + \frac{J_R}{J_I} (i \omega \tau_1)^{-1} \right]$$

(3)

with $J_I$ and $J_R$ : instantaneous and relaxed compliance respectively, $\tau_1 = \frac{1}{C_2}$, $\tau_2 = \frac{1}{C_1 C_2}$ and $\gamma$: constant dependant on the width of the distribution ($0 < \gamma < 1$).

The relation (3) is really compatible with experimental results as shown in figures 1 and 2 where theoretical $\tan \phi$ is calculated in function of $\omega$ with the relation (3).

In order to describe the creep curves, the function $J(t)$ obtained from (3) must be used. For short times, $J(t)$ is given by:

$$J(t) = J_I \exp \left( \frac{-\tau_2}{\tau_1} \right) + \frac{\frac{\gamma}{n}}{n}$$

(4)

with $n$: viscosity.

Once more, it can be verified that the equation (4) is compatible with experimental results as shown in figure 3.

To sum up, the assumption about the shear microdomains (smd) induced by the applied stress, which was previously considered to interpret only the plasticity of metallic glasses, seems in agreement with the double aspect of the behaviour of glassy materials near $T_g$ (viscoelastic and viscoplastic). Furthermore, although the rate of formation of smd varies linearly with stress, a non newtonian flow of glasses is observed with high values of this stress: a more realistic function $f(\sigma_i)$ (relation (2)) would be able to describe this feature.

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