Deformation of Al$_{85}$Y$_8$Ni$_5$Co$_2$ Metallic Glasses under Cyclic Mechanical Load and Uniform Heating

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Abstract: Modelling of the deformation process of Al$_{85}$Y$_8$Ni$_5$Co$_2$ amorphous alloys was carried out under simultaneous application of cyclic mechanical load (at 0.3 or 3 Hz frequencies) and continuously increasing temperature (heating rate 5 K/min). It is shown that deformation of the amorphous specimens occurs by the hyperbolic temporal dependence. It is analytically determined and experimentally proved that for non-isothermal cyclic deformation, the wave effects take place as a result of the superposition of thermal-activated and mechanical components. The behaviour of the material under thermo-mechanical action was described qualitatively within the framework of Spaepen’s model. The dependencies for the reaction force of the samples were obtained as two-parameter functions of the frequency and temperature. A reaction force surface of a specimen, as a function of the different forcing frequencies and time, has been plotted.

Keywords: metallic glasses; mechanical properties; inorganic materials; thermal expansion

1. Introduction

The nature of glass transition process is one of the main unsolved problems of solid-state physics [1]. Under fast enough cooling of the metallic melt, amorphous metallic alloys (metallic glasses, MG) undergo glass transition. They exhibit the absence of the long-range order in their atomic structure. This type of metallic alloys is an object of persistent investigations [2–6] from the moment of their first synthesis [7]. Metallic glasses show advanced mechanical [8], magnetic [9] and other properties. Aluminium-based metallic glasses are perspective materials due to their high tensile strength, lightness, and, consequently, specific strength [10,11]. On heating, these materials undergo nanocrystallization formation of primary aluminium [12–16] that leads to their additional hardening. The process of structural relaxation precedes crystallization [17].

An investigation of the mechanical deformation of Al$_{85}$Y$_8$Ni$_5$Co$_2$ amorphous alloy on heating with additional cyclic loading is an interesting research task. The mentioned impact is one of the widespread pre-treatments such as isothermal annealing [18] and chemical processing [19], which are used at the technology of different materials. The results in this field can broad understanding of the physical processes, related to β-relaxation [20] and hardening because of a strong impact of loading and temperature on continual structure. Moreover, isothermal creep is well investigated both theoretically [21] and experimentally [22] right now, but there are few works on modelling of deformation in metallic glasses at non-stationary conditions (for example, at dynamic mechanical analysis—DMA).
Thus, the model description of the DMA experiments can help conveniently with a generalization of some important experimental cases.

2. Materials and Methods

The ingots of Al$_{85}$Y$_8$Ni$_5$Co$_2$ alloy were made by the arc melting of pure components (99.9 wt.% purity) in an argon atmosphere (Ti was a gas absorbent). We made the amorphous ribbons at about 40 µm in thickness and 4.5 mm in width by the melt spinning method. The produced specimens were placed between two steel plane plates with 1-mm thickness, and then exposed to cold rolling with the tangent velocity of 0.628 m/s to decrease the initial thickness by 35 ± 5%. The amorphous structure was studied by the X-ray diffractometry with Cu-K$_\alpha$ monochromatic radiation. Thermal stability of the specimens was verified by differential scanning calorimetry at the heating rate of 5 K/min. Deformation of the specimens with 30-mm length and 18.5-mm working zone (width ~4.5 mm, thickness ~0.026 mm) was carried out using a Q800 dynamic mechanical analyser (TA Instruments) on heating up to 540 K and a rate ~5 K/min. Before testing, the specimen had been loaded by the constant stress of 20 MPa that was fixed until the end of the experiment. In addition to the constant stress, the specimen was also loaded in the sinusoidal mode at 20-kPa amplitude and different frequencies of 0.3 and 3 Hz.

3. Results and Discussion

The glass-transition and primary crystallization temperatures for the Al$_{85}$Y$_8$Ni$_5$Co$_2$ amorphous alloy were found to be 533 and 545 K, respectively (Figure 1).

![Figure 1. Thermogram of Al$_{85}$Y$_8$Ni$_5$Co$_2$ glassy alloy.](image-url)

All specimens have amorphous structure that was confirmed with X-ray diffractometry (Figure 2).

When tested under the cyclic load, the specimen elongates at a higher velocity compared to that at the constant load. The increase in the oscillation frequency from 0.3 to 3 Hz leads to a further increase in the deformation velocity of the specimens.
Deformation of the Al<sub>85</sub>Y<sub>8</sub>Ni<sub>5</sub>Co<sub>2</sub> metallic glass specimen at the 0.3 and 3 Hz load frequencies is plotted in Figure 3a.

It is interpolated with a high accuracy by the hyperbolic function [23] (the «model» curve):

$$l(t) = l_0 + \frac{Ct}{B^2 - Bl}$$  \hspace{1cm} (1)
where $l_0$—is initial length; $C$ and $B$—are analytical fixed parameters having (m·s) and (s) units, respectively. $B$—this parameter is time of the fracture of a specimen; $t$—current deformation time.

The analytical relation (1) is more applicable for the description of the experiments in comparison with superposition of the exponential functions (from one to three terms). Herewith, the whole deformation process, with account for thermal expansion (the value of linear coefficient of thermal expansion—$\alpha$ is insignificant after $\Delta l = 6.4 \times 10^{-5}$ mm at $t \geq 5.7$ min), is described by one equation (hyperbola) with the correlation coefficient of more than 0.99 and with clear physical meaning of the functional parameters. As the heating rate was held constant, the temperature is linked to time by the following relation:

$$ \frac{t}{V_T} = \frac{T - T_0}{V_T} $$

(2)

where $T_0$ and $T$—the initial and current temperatures of heating; $V_T$—the heating rate. As for accuracy, dimension, and physical meaning retain in Equation (1), after its differentiation and other mathematical operations [23], the proposed hyperbolic relation can be also used for the further modelling of the experiment.

In this case, the specimen can be considered as a continual spring (Figure 3b) that acts on the moving grip (material point) of the DMA-machine with a reaction force, because of action of the external load on the same material point from the testing machine. A spatial position of the material point, whose deviation depends on the reaction force of the spring, is described by the $x = l(t)$ function. For this case, Newton’s second law of motion, projected on the $x$-axis (deformation axis) is represented as:

$$ m\ddot{x} = F_{\text{load}} - A \sin(\omega t) - F_{\text{reaction}} $$

(3)

where $m$—is mass of the material point (the machine grip), $\ddot{x}$—acceleration of the material point, $F_{\text{load}}$—constant external load from the testing machine, $A$—amplitude of external additional (load) oscillations, $\omega$—angular frequency of oscillations ($2\pi \cdot 3$ Hz equals $6\pi$ rad/s), and $F_{\text{reaction}}$—reaction force of the amorphous specimen (the spring). Projections of the law of motion on other axes are equal to zero. In compliance with (2) and also with relation for strain $\varepsilon$:

$$ \varepsilon = \frac{x(t) - x_0}{x_0} = \frac{Ct}{x_0(B^2 - Bt)} = \frac{C(T - T_0)}{x_0B(V_T + T_0 - T)} $$

(4)

Equation (3) can be rewritten in relative magnitudes:

$$ m\ddot{x}_0 = F_{\text{load}} - A \sin(\omega t) - F_{\text{reaction}} = F_{\text{load}} - A \sin(\omega \frac{T - T_0}{V_T}) - F_{\text{reaction}} $$

(5)

where $x_0$ is the initial length of the specimen and $\ddot{x}$ is the acceleration as a relative magnitude. The transformation of (1) to the relative form (4) (without equation (2) or with its substitution instead of $t$ value), with the further double-time differentiation of (4) and substitution of the second-order derivative into (5) instead of $\ddot{x}$, permits one to find the analytical form for the reaction force of the specimen—$F_{\text{reaction}}$, as a function from frequency and deformation time (or temperature):

$$ F_{\text{reaction}}(\omega, t(T)) = F_{\text{load}} - A \sin(\omega t) - \frac{2mc}{(B - t)^3} = $$

$$ F_{\text{load}} - A \sin(\omega \frac{T - T_0}{V_T}) - \frac{2mcV_T^3}{(BV_T + T_0 - T)^3} $$

(6)

For an additional evaluation of the applicability of Equation (6) to the experiment, analysis of changing of this function is necessary within the range of validity of Hooke’s law (at $t \to 0$, $T \to T_0$ and $\Delta x \to 0$). After the single-valued solving of Equation (4) for deformation
time and substitution of the received equation into (6)—instead of \( t \), it is possible to determine reaction force as a function of \( \Delta x \) value:

\[
F_{\text{reaction}} = F_{\text{load}} - A \sin\left(\frac{\omega B^2 \Delta x}{C + B \Delta x}\right) - \frac{2mC}{(B - \frac{B^2 \Delta x}{C + B \Delta x})^3}
\] (7)

In the received Equation (7), we transform the third term (reducing a fraction to a common denominator) and also the sinusoidal function (\( -1 \leq \sin(\bullet) \leq 1 \Rightarrow \sin(\bullet) \sim \pm 1 \)), and then, after neglecting of the smaller \( (\Delta x)^3 \) and \( (\Delta x)^2 \) quantities, we have finally a relation for reaction force:

\[
F_{\text{reaction}} \approx F_{\text{elastic}} = F_{\text{load}} \mp A - \frac{6m}{B^2 \Delta x} - \frac{2mC}{B^3}
\] (8)

From the relation (8), it can be noticed that the terms without the \( \Delta x \) value have the dimension of force—\([N]\), and the multiplier at \( \Delta x \) has the dimension of the elastic coefficient—\([kg/s^2]\). Consequently, Equation (8) can be also presented in a form of \( F_{\text{elastic}} - Q \approx -k \Delta x \), that is a particular case of Hooke’s law. Thus, the proposed relation (6) is suitable because it describes analytically a transition from reaction force to Hooke’s law at the small deformations. The functional relation between the elastic coefficient \( k \) of a specimen and its fracture time \( B \) is estimated only qualitative from Equation (8) as \( k \approx \frac{6m}{B^2} \).

The value \( k \approx \frac{6m}{B^2} \) taken from Equation (8) allows for estimating (only qualitative) of the functional relation between the elastic coefficient of a specimen and its fracture time at the presence of plastic deformation.

For an analytical description of a particular non-isothermal experiment with a fixed frequency (3 Hz) of the mechanical load, we consider the function (6) in its relative forms with time in minutes:

\[
\frac{F_{\text{reaction}}(\omega \neq 0)}{F_0} = 1 - 0.0735 \sin(6\pi t) - \frac{0.01}{(52.435 - t)^3} = 1 - 0.0735 \sin(0.365T - 109.5) - \frac{202.7}{(851.2 - T)^3}
\] (9)

\[
\frac{F_{\text{reaction}}(\omega = 0)}{F_0} = 1 - \frac{0.01}{(52.435 - t)^3} = 1 - \frac{202.7}{(851.2 - T)^3}
\] (10)

that are particular cases of Equation (6) with fixed parameters of the experiment \( (F_0) \)—the initial magnitude of reaction force). The plots of the received functions have been shown in Figure 4.

As seen in Figure 4, when the cyclic load increases, reaction force has the lower magnitudes, as an opposite from the reaction force with zero frequency of the external load, which explains the reason of acceleration of deformation in the \( \text{Al}_{85}\text{Y}_{8}\text{Ni}_{5}\text{Co}_{2} \) amorphous specimens on cyclic loading. Periodical impact of the external load with fixed non-zero frequency sufficiently reduces the elasticity of the material, similarly to the fatigue deformation process. Herewith, in frames of the calculating and the beating, whose occurrence can be a result of the impact of several vibrational modes in whole deformation process, has been observed on the force curve with non-zero frequency. Particularly, the beating can be a result of mutual overlapping of the processes activated by thermal deformation and frequency loading—the thermal and mechanical modes. The occurrence of the mentioned deformation processes also can lead to the appearance of interference and resonance in the material. Herewith, the dynamics of the thermal part is described by the hyperbolic term in (6), and the frequency part is described by the sinusoidal term in the same equation. The relations of the reaction force depicted in Figure 4 are comparable with the experimental relation of the resultant force (Figure 5). For elimination of the initial dilatometric expansion of the specimen at the heating and the data of the DMA-machine stabilization in the load regime, the curve is plotted in Figure 5, beginning at 12 min.
When the application of the external load of a 3-Hz frequency coupled with additional thermal deformation and frequency loading occurs, resultant frequency—because of the continuity of the specimen. Frequency of the external load, which explains the reason of acceleration of deformation in the Al_{85}Y_{8}Ni_{5}Co_{2} amorphous specimen, depending on viscoelastic properties of the material (elastic coefficient, etc.).

A graphical elimination of the initial dilatometric expansion of the specimen at the heating and the load oscillations included in the argument of the sinusoidal in the calculations (Equation (6)), resonance occurs on the experimental curve. It is known that the beating can be a result of the mutual overlapping of the processes activated by various vibrational modes in whole deformation process. Herewith, the dynamics of the thermal part is described by the hyperbolic term in (6), and the frequency part is described by the sinusoidal term in the same equation. The relations of the reaction force depicted in Figure 4 are comparable with the experimental relation of the resultant force (Figure 5). For this process, the difference between model and the experiment is given by the impossibility on account of the additional increase of oscillation frequency in the model and experimental curve differ from frequency of the external load. It can be a result of heating and viscoelasticity of the specimen (the difference of frequency in the thermal deformation process, has been observed on the force curve with non-zero frequency. Particularly, the beating can be a result of the impact of several vibrational modes in whole deformation process. Herewith, in frames of the calculating and the beating, whose occurrence can be a result of the impact of several vibrational modes in whole deformation process. Herewith, in frames of the calculating and the beating, whose occurrence can be a result of the impact of several vibrational modes in whole deformation process. Herewith, in frames of the calculating and the beating, whose occurrence can be a result of the impact of several vibrational modes in whole deformation process. Herewith, in frames of the calculating and the beating, whose occurrence can be a result of the impact of several vibrational modes in whole deformation process.

Thus, in Figure 5, the graph of a sum of the external oscillating load and reaction force of an amorphous specimen, depending on viscoelastic properties of the material (elastic coefficient, etc.). A graphical elimination of the initial dilatometric expansion of the specimen at the heating and the load oscillations included in the argument of the sinusoidal in the calculations (Equation (6)).
Comparison of the data in Figures 4 and 5 shows that the calculated values of reaction force decrease slower about experimental relation, that is only qualitative accordance. However, the oscillation amplitudes depicted in Figures 4 and 5 decrease with time, and the beating is also observed on the experimental curve. The oscillation frequencies in the model and experimental curve differ from frequency of the external load. It can be a result of heating and viscoelasticity of the specimen (the difference of frequency in the calculated curve distinctly relates to the presence of the hyperbolic term in Equation (6)). Herewith, for the experimental curve (Figure 5), an increase of oscillation frequency in time is typical, that is not predicted by the mentioned model equations. In the case of the cyclic load, the hardening replacing by the unloading occurs in the specimen periodically, i.e., the energy of the elastic deformation with the further jump-like unloading accrues. This process accelerates with temperature. For this process, the difference between the model and the experiment is given by the impossibility on account of the additional functional relation between natural oscillation frequency of the specimen and temperature—because of basic hypothesis about the continuity of the specimen. Frequency of the load oscillations included in the argument of the sinusoidal in the calculations (Equation (6)) is the fixed value that is set in the testing machine at the beginning of the experiment. Thus, in Figure 5, the graph of a sum of the external oscillating load and reaction force of specimen is shown taking into account the thermo-activated natural oscillations.

When the application of the external load of a 3-Hz frequency coupled with additional response from the specimen with frequency $\omega(T)$, including the reaction force of the specimen (Equation (6)), resonance occurs on the experimental curve. It is known from the theory of wave process that while superposition of two harmonic oscillations with the frequencies $\omega_1$ and $\omega_2$ occurs, resultant frequency $\omega_{\text{result}}$ is determined from the relation $|\omega_1 - \omega_2| = \omega_{\text{result}}$, where (for our case) $\omega_1 = 6\pi \text{ rad/s}$—is frequency of the external load, $\omega_2 = \omega(T)$—natural oscillation frequency of an amorphous specimen, depending on viscoelastic properties of the material (elastic coefficient, etc.). A graphical calculation of resultant frequency $\omega_{\text{result}}$ in Figure 5 permits one to find the natural oscillation frequency of a specimen during the whole deformation process. As the heating temperature increases, natural oscillation frequency of a specimen decreases because of the decrease of the stiffness that gives an increase of resultant frequency at constant frequency of the external load. A force jump occurs in Figure 5 (at 45–47 min), probably, due to tuning of the natural oscillation of the specimen to the resonance magnitude. Herewith, $\omega_2/\omega_1 \approx 0.97$ near the time of the jumping (47 min). That is comparable with the necessary condition of equality between the external and the natural frequencies at resonance. Thus, the proposed model has predicted clearly the existence of the beats and resonance together with the general aspiration to zero the reaction force of the material, that can be used in the future for investigation of the mechanical deformation in MGs, on heating at cyclic load.

Acceleration of the deformation under cyclic load also can be explained in frames of the Spaepen’s free volume model [24]. Under a cyclic load, decrease of the potential barrier is a probable process, that gives an accelerated transition of the system to a collective atomic shift (to the plastic deformation) [24]. In this case, as opposed to the case of the constant external load, the free energy in a normal-distributed atomic system of MG is changed. Herewith, the free energy of the MG system not only cannot be a constant, but it can decrease on heating under cyclic load leading to faster deformation of the material. The probability of overcoming of the potential barrier by more and more number of atoms, those shift along the direction of cycling load is increased. The accelerated collective atomic shift (growth of average free volume in a material) leads to a noticeable increase in the deformation rate on the experimental curves at a higher frequency of the external load.

For the definitive analysis of the different conditions of the mentioned oscillating system, it is necessary to consider the general view of the reaction force surface that is given by Equation (6) and plotted in Figure 6.
As can be seen from the force surface (Figure 6), the occurrence of the resonance or interferential areas [25], which is similar to Chladni figures in acoustics [26], takes place in the case of the studied metallic glass under cyclic mechanical loading on heating (2). The beating depicted in Figure 4 is a special case of the general force surface, and, consequently, it is a part of wide-scale wave processes.

4. Conclusions

The resonance processes observed in the Al$_{85}$Y$_5$Ni$_5$Co$_2$ metallic glass are caused by the superposition of thermal activation and cyclic loading. It leads to faster deformation of a specimen at a fixed frequency. Herewith, the occurrence of the wave areas (nodes and antinodes), wherein elastic force is constant (regions of periodic stability), and is found to be an essential behaviour of the specimen.

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