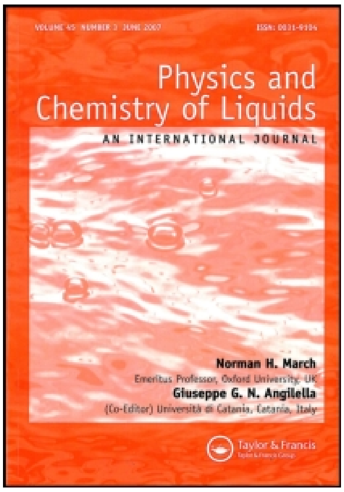

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Viscosity of glass forming $Al_{86}Ni_8(La/Ce)_6$, $Al_{86}Ni_6Co_2Gd_4(Y/Tb)_2$ melts

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The temperature and time dependences of kinematic viscosity in $Al_{86}Ni_8(La/Ce)_6$, $Al_{86}Ni_6Co_2Gd_4(Y/Tb)_2$ melts over the range of liquidus up to 1300°C have been investigated. An irreversible non-monotonic change of the melts viscosity above the temperature of melting brought about by the destruction of their microheterogeneous state inherited from the multi-phase solid sample has been found out. It is shown that for the melts transition into the quasi-equilibrium state long isothermal holding is necessary. The relaxation time decreases with the increase of the melt temperature.

Keywords: Melt; Viscosity; Hystereses; Relaxation; Non-equilibrium microheterogeneous state

1. Introduction

In recent years amorphous Al-based alloys (80–90 at.%) with transition (TM) and rare-earth metals (REM) have increasingly attracted interest. Rapid solidification of aluminium alloys of the Al-TM-REM type makes it possible to produce a new generation of aluminium alloys with higher strength, plasticity, corrosion resistance as well as with increased thermal expansion and wear resistance [1,2] in comparison with the properties of cast commercial alloys.

Increasing the glass-forming ability and stability of the physical–mechanical properties of the amorphous alloys is connected with the possibility of their structure effect on their formation during the process of superfast quenching of the melt due to the optimisation of the technological conditions of their production including the temperature and time of holding in the liquid state.

Choosing the conditions of thermal treatment and time, it is necessary to take into account the possibility of structure transformations and relaxation processes in the melt, whose indicator is the temperature and time dependences of structure-sensitive

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properties, in particular, viscosity [3]. Besides this, after melting a multi-component alloy may have a non-equilibrium microheterogeneous state, which results in decreasing the glass-forming ability of the melts [4], the establishment of equilibrium in the melt sometimes last for hours, which is registered in the time dependences of these properties [5].

The temperature and time dependences of kinematic viscosity of the $Al_{86}Ni_8(La/Ce)_6$, $Al_{86}Ni_6Co_2Gd_4(Y/Tb)_2$ melts have been investigated in the present article. The peculiarities of the relaxation processes in these melts have been studied.

2. Experimental procedure

The samples under study were melted in an arc furnace in the atmosphere of purified argon after preliminary vacuum pumping of the working chamber up to 10^{-5} mm of the mercury column. The initial components for melting the alloys were the elements with the content of the basic metal: aluminium 99.99; lanthanum 99.99; yttrium 99.8; cerium 99.9; gadolinium 99.9; terbium 99.9; electrolitical nickel 99.96 and cobalt 99.96 mass%.

Measuring the kinematic viscosity (ν) of the liquid alloys was carried out by the method of damped torsional vibrations of the cylindrical crucible with the melt [6]. The determination of the logarithmic decrement (δ) and period of vibrations of the suspended system of viscosimeter (τ) was carried out by the optical method using helium–neon laser and a precision photosensor.

Calculating the viscosity with the help of numerical methods [7], equation was solved:

$$f(\nu) = \mathrm{Re}(L) + \frac{\delta}{2\pi}\mathrm{Im}(L) - 2I\left(\frac{\delta}{\tau} - \frac{\delta_0}{\tau_0}\right) = 0, \tag{1}$$

where I – the inertia moment of the suspended system;

δ_0 and τ_0 – the decremeter of damping and period of vibratious of the suspended system without a sample;

$\mathrm{Re}(L)$ and $\mathrm{Im}(L)$ – real and imaginary parts of the friction function L [6].

From equation (1) viscosity cannot be expressed explicitly, which complicates the error estimation in its calculation. To calculate the error, a relative change of viscosity caused by the error of each experimentally determined value, while other parameters were unchanged, was determined. In particular, a mean square error in the calculation of the temperature dependence of viscosity due to the change of δ and τ, is calculated by:

$$\left(\frac{\Delta\nu}{\nu}\right)_T = \left[\left(\frac{\Delta\nu}{\nu}\right)^2_{\Delta\delta} + \left(\frac{\Delta\nu}{\nu}\right)^2_{\Delta\tau}\right]^{1/2}$$

where $(\Delta\nu/\nu)_{\Delta\delta}$ and $(\Delta\nu/\nu)_{\Delta\tau}$ – are relative deviations of viscosity on changing δ and τ within their errors. A total mean square error in the viscosity calculation was estimated according to the formula:

$$\left(\frac{\Delta\nu}{\nu}\right)_\Sigma = \left[\left(\frac{\Delta\nu}{\nu}\right)^2_T + \sum_n \left(\frac{\Delta\nu}{\nu}\right)^2_{\Delta n}\right]^{1/2},$$

where n are all the values calculated in equation (1) and friction function: the moment of inertia of the suspended system, mass, radius and height of the sample. The total mean square error in the measurement of ν does not exceed 4% with the error in a single measurement being not higher than 2.5%.

Prior to measuring the alloys were re-melted directly in the viscosimeter for not less than 1 h at the temperatures by 300–400°C increasing their temperatures of melting. All the measurements were carried out in the atmosphere of high purity helium. Cylindrical Al_2O_3 crusibles with an inner diameter of 16 mm and height of 40 mm were used. To eliminate the effect of the oxide film formed on the alloy surface, an Al_2O_3 cover was placed above the sample, which was used as a second face surface. The construction of the cover allowed it to move freely along the vertical crucible axis and rotate during torsional vibrations together with it. The temperature in the viscosimeter furnace was determined with the W-Rh(5%)–W-Rh(20%) thermocouple with the precision of ±10°C and it was maintained on the same level with the help of a high-precision temperature control (HTR) with the precision of ±0.5°C.

3. Results and discussion

Figure 1 gives characteristic temperature dependences of viscosity of the $Al_{86}Ni_8La_6$, $Al_{86}Ni_6Co_2Gd_4Tb_2$ melts, obtained under heating and following cooling after isothermal holdings for 1800 and 300 s at each temperature. Under heating a relatively sharp decrease of viscosity and its hysteresis under the following cooling are observed. In the polytherm of the $Al_{86}Ni_8La_6$ melt viscosity obtained under heating with isothermal holdings at each temperature for 1800 s (curve 1, figure 1a) one can observe the maximum in the vicinity of 890°C on the background of a relatively sharp decrease of ν values over the temperature range from t_L to 970°C. On cooling the melt one can see the hystereses of viscosity below 970°C that remains up to its crystallization. Decreasing the time of holding at each temperature down to 300 s (curve 2, figure 1a) results in the shift of the maximum in the $\nu(t)$ dependence under heating and that of the temperature of the beginning of the viscosity hysteresis towards higher temperatures: 1130 and 1250°C, respectively (figure 1a). In the viscosity polytherm of the $Al_{86}Ni_6Co_2Gd_4Tb_2$ melt under heating with isothermal holdings at each temperature during 30 min (curve 1, figure 1b) over the temperature range from t_L up to 970°C one can also see a relatively sharp viscosity decrease followed by hysteresis under cooling. The decrease of the holding time at each temperature leads to the increase of the temperature of the hysteresis beginning up to 1270°C (curve 2, figure 1b). The viscosity polytherms of the $Al_{86}Ni_8Ce_6$, $Al_{86}Ni_6Co_2Gd_4Y_2$ melts have a similar shape.

Under cooling the temperature dependences for all the melts studied are well-described by the Arrhenius equation (figure 2):

$$\nu = A \cdot \exp\left(\frac{E}{RT}\right), \qquad (2)$$

where A is a constant value;

E is the energy of the viscous flow activation;
R is the molar gas constant;
T is the absolute temperature.

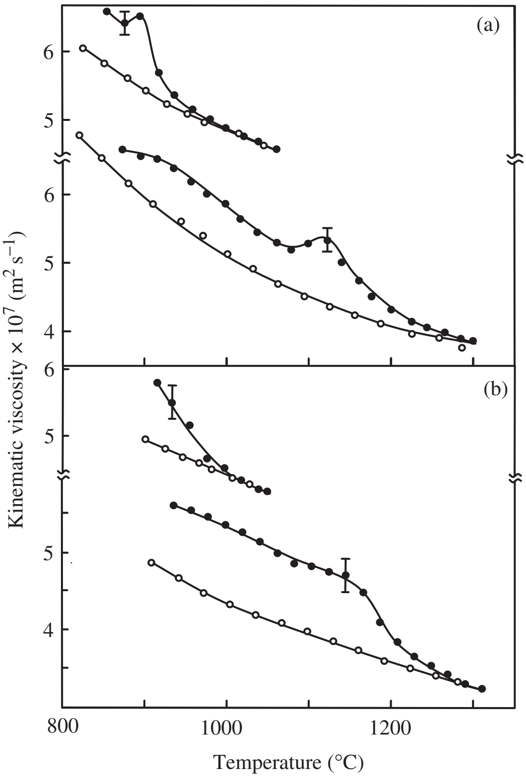

Figure 1. Temperature dependences of the viscosity in $Al_{86}Ni_8La_6$ (a) and $Al_{86}Ni_6Co_2Gd_4Tb_2$ (b) melts obtained under the condition of isothermal holdings at each temperature during 1800 (curves 1) and 300 (curves 2) s: ● – heating; ○ – cooling.

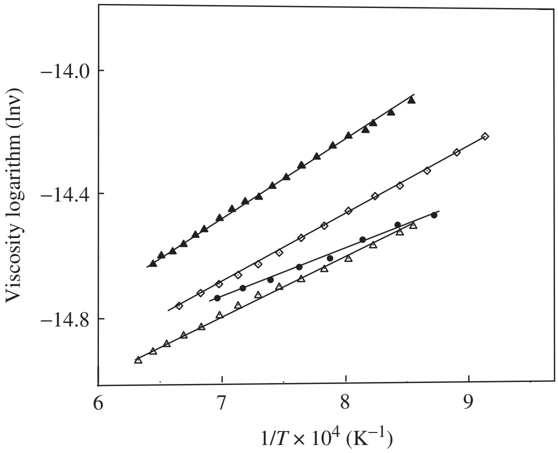

Figure 2. Temperature dependences of the viscocity in $Al_{86}Ni_8La_6$ (◇), $Al_{86}Ni_8Ce_6$ (●), $Al_{86}Ni_6Co_2Gd_4Tb_2$ (▲), and $Al_{86}Ni_6Co_2Gd_4Y_2$ (△) under cooling (logarithmic coordinates).

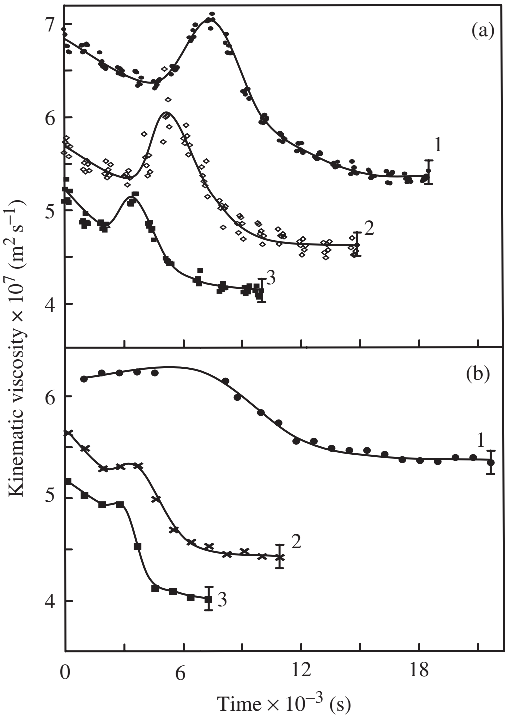

Figure 3. Time dependences of the $Al_{86}Ni_8La_6$ melt (a) at the temperatures 900°C (1), 1000°C (2) 1100°C (3) and those of $Al_{86}Ni_6Co_2Gd_4Tb_2$ (b) melt at the temperatures 920°C (1), 1100°C (2), 1200°C (3) after heating from room temperature.

To find out the reasons for viscosity hysteresis, we studied the temperature dependences of ν in melts under different overheating above the liquidus temperature during long isothermal holdings. The given temperature was reached at the velocity of 30–40°min^{-1}. Figure 3 presents the time dependences of the $Al_{86}Ni_8La_6$ (figure 3(a)) viscosity at 900, 1000 and 1100°C and $Al_{86}Ni_6Co_2Gd_4Tb_2$ melts at 920, 1100 and 1200°C under isothermal conditions after heating from the room temperature. All the curves obtained have a non-monotic character: on the background of decreasing viscosity dependence one can observe the maximum as time goes by. The relative value of viscosity and the time at which the maximum reveals itself, decrease with the increase of the overheating temperature of the melt with respect to liquids.

According to the time dependences obtained the relaxation times of viscosity (τ_Γ) in the melts under different overheating conditions above liquidus were determined. τ_Γ was determined as the time period during which in the course of isothermal holding constant viscosity values are established within the experimental error. The relaxation times of viscosity in all melts studied at different temperatures are given in figure 4 (in semilogarithmic coordinates). From figure 4 it is seen that the values of τ_Γ for different melts at similar temperatures coincide within the experimental error and the temperature dependence of τ_g is exponential.

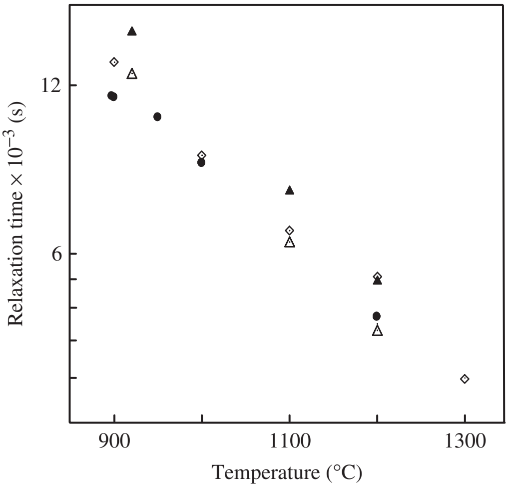

Figure 4. Time dependences of relaxation time of the viscosity τ_r in the $Al_{86}Ni_8La_6$ (◇), $Al_{86}Ni_8Ce_6$ (●), $Al_{86}Ni_6Co_2Gd_4Tb_2$ (▲) and $Al_{86}Ni_6Co_2Gd_4Y_2$ (△) melts on the temperature (semilogarithmic coordinates).

Numerous diffraction studies of metal melts, Al-based alloys included, show their complex microheterogeneous structure (e.g., [8]). Non-equilibrium microheterogeneous state of the liquid alloys investigated in the present work is due to the availability of atoms microgroups with the ordering according to the type of initial crystalline phases in them (in particular, microgroups based on high-melting chemical compounds Al_3Ni, $Al_{11}La_3$, $Al_{11}Ce_3$, Al_3Y). The solution of the latter under the effect of temperature and time of holding, results in an irreversible change of the melt structure and its gradual approach to the equilibrium state, which is reflected in the relaxation of structure-sensitive properties of the melts, included viscosity. Increasing the temperature of the melts up to 1300°C (overheating above liquidus of more than 400°C) does not reveal the threshold temperature, but only reduces the relaxation time (τ_r) towards equilibrium.

Non-monotonicity of time dependences of the melts viscosity is due evidently, to non-uniform dissolution and dispersion of non-equilibrium microgroups.

4. Conclusions

The temperature and time dependences of kinematic viscosity in the $Al_{86}Ni_8La_6$, $Al_{86}Ni_8Ce_6$, $Al_{86}Ni_6Co_2Gd_4Y_2$, $Al_{86}Ni_6Co_2Gd_4Tb_2$ melts have been investigated over the range from t_L to 1300°C. Irreversible changes of viscosity under overheating of the melts above the temperatures of liquids due to the decomposition of the microheterogeneous state of the melts inherited from the multi-phase solid samples have been found out. It is shown that for the melts to transform into the equilibrium state long isothermal holdings are necessary, the increase of the melt temperature results in the decrease of its relaxation time.

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