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THE TEMPERATURE DEPENDENCE OF COEFFICIENT OF VISCOSITY FOR $Cu_{60}Zr_{40}$ AND $Ni_{64}Zr_{36}$ METALLIC GLASSES

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The cold finger method was used for dilatometric measurements of $Cu_{60}Zr_{40}$ and $Ni_{64}Zr_{36}$ amorphous samples at elevated temperatures under applied tensile stress. The creep process was identified from the experimental data by separation of the free volume annealing process. It was found out that this process occurs at lower temperatures for the $Ni_{64}Zr_{36}$ system and that it follows a linear time dependence. The coefficient of viscosity was evaluated from the data for creep for both systems, their functional dependence on temperature was given and activation energies for the systems were evaluated. The obtained temperature dependence of the coefficient of viscosity was of the form $\eta = \eta_0 \exp(\Delta E/kT)$. The evaluated activation energies for $Cu_{60}Zr_{40}$ and $Ni_{64}Zr_{36}$ amorphous systems were 178.8 kJ/mole and 41.2 kJ/mole, respectively.

1. Introduction

The cold finger method is a modification of dilatometric investigation of amorphous samples[1]. The method is very suitable for investigation of dilatometric changes during the isothermal treatment and enabled us to obtain information about the activation energies which define the process at a fixed temperature. The analysis of the results obtained by dilatometric measurements enabled us to separate the processes which occured during the isothermal treatment. There are two processes which are dominant in the amorphous samples: a) the sample contraction due to the loss of the excess free volume and b) the sample extention influenced by the creep process. The process of contraction influenced by annealing of the excess free volume follows a logarithmic law[2]. The separation of the two mentioned processes can be done graphically by adapting through fitting the first values to the logarithmic dependence of the type

$$\frac{\Delta l}{l} = B \ln t + A,$$

where A and B are constants. This relation describes the specific case of the lenght change behaviour due to the loss of the free volume; it comes from the dependence which can be applied to all parameters which may vary in the case of the free volume change.

At the beginning of the process the experimental values follow this dependence very closely. As the creep process becomes more dominant, this dependence is no more followed, so the contribution of the creep process can easily be defined.

It can be shown that the length change of the sample due to the creep is a linear function of time. The direction coefficient ($\dot{\varepsilon}$) of that dependence gives us the length change per time unit. When $\dot{\varepsilon}$ is known it is easy to define the coefficient of viscosity[3] by the use of the relation

$$\eta = \frac{1}{3} \frac{\sigma}{\dot{\varepsilon}},$$

where σ is the applied tensile stress.

In this paper the method of separation was used for paralel investigation of two amorphous systems: $Cu_{60}Zr_{40}$ and $Ni_{64}Zr_{36}$.

2. Experimen and results

The amorphous $Cu_{60}Zr_{40}$ and $Ni_{64}Zr_{36}$ samples were in form of thin ribbons 17 mm long, 2.004 mm wide and 0.030 mm thick. They were obtained from the melt by the rotating wheel method. The length of the samples was measured for normalization to the same length by use of a travelling microscope. The thickness was measured by a micrometer with an accuracy of $\pm$ 0.005 mm.

TABLE 1

The sample	T_g (K)	T_1 (K)	T_2 (K)
$Ni_{64}Zr_{36}$	836	864	885
$Cu_{60}Zr_{40}$	763	917	922

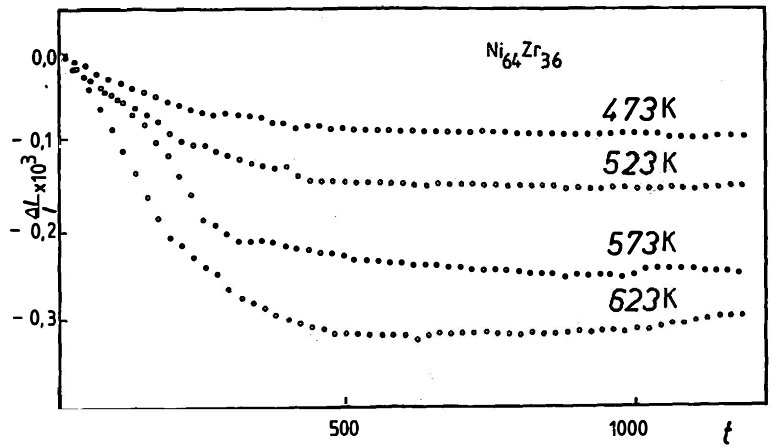

Fig. 1. The length change of amorphous $Ni_{64}Zr_{36}$ sample in the temperature range from 473 K to 623 K. The applied tensile stress was 116.30 kPa.

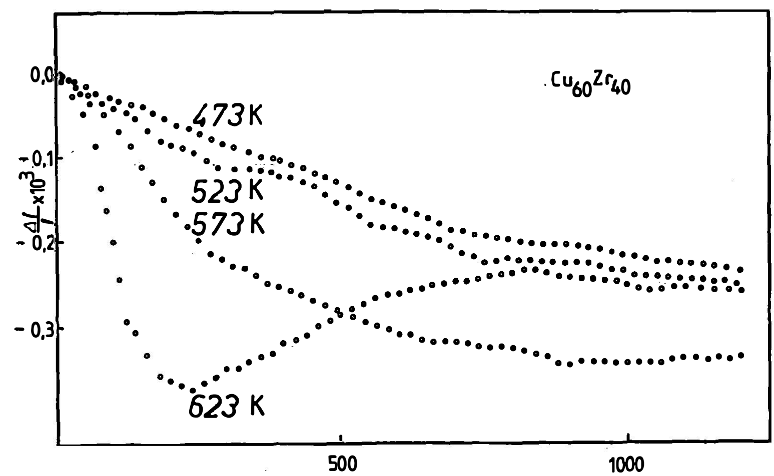

Fig. 2. The length change of the amorphous $Cu_{60}Zr_{40}$ sample in the temperature range from 473 K to 623 K. The applied tensile stress was 116.30 kPa.

The experiments were performed on as quenched samples in a temperature range from 473 K to 623 K. The temperature of the isothermal treatment was measured with an accuracy of $\pm$ 0.01 K[1]. This temperature range is within the domain where the crystallisation process occurs. The characteristic glass temperature (T_g) and crystallisation temperatures (T_1 and T_2) at heating rate of 80 K/min are given in Table 1[4]. The temperatures T_1 and T_2 are related to the two phases of the crystallisation process. The experimental data for the both systems are given on Fig. 1 and Fig. 2. In all experiments we have used the same value of the applied tensile stress which was 116.30 kPa; this was necessary for the investigation of the coefficient of viscosity. All the obtained results were not drown because of the overlap of the curves.

The graphical separation for both systems is shown in Fig. 3 and Fig. 4. Identical treatment was used for each curve i. e. for all the obtained experimental data. The experimental results are given by empty circles and the full points show

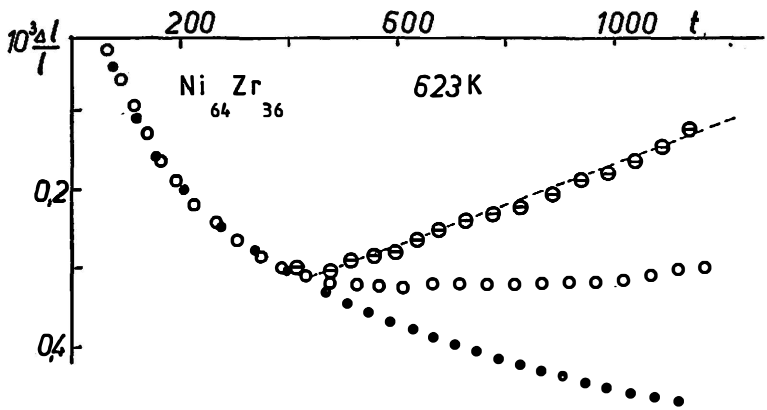

Fig. 3. The process of the graphical separation of the creep process for $Ni_{64}Zr_{36}$ amorphous sample at 623 K. The empty circles represent the experimental values and the full points are the fitted values. The creep process is shown by the circles with a hyphen.

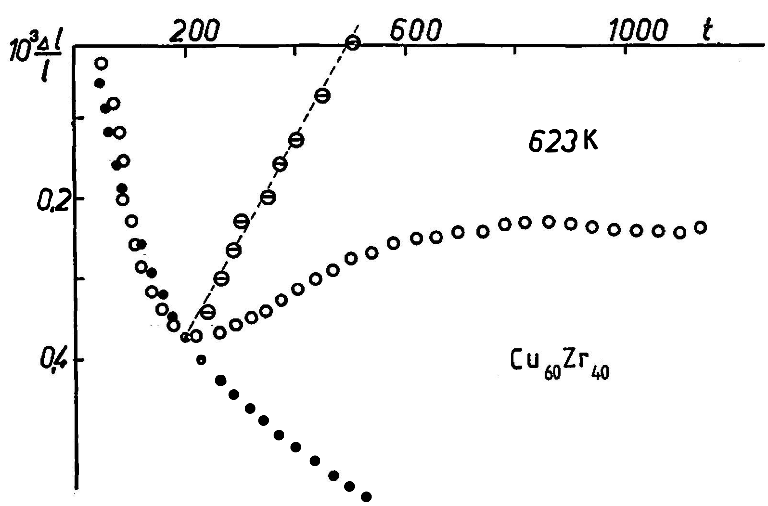

Fig. 4. The process of the graphical separation of the creep process for $Cu_{60}Zr_{40}$ amorphous sample at 623 K. The empty circles represent the experimental values, the full points are the fitted values and the creep process is shown by circles with a hyphen.

the values obtained by fitting on the basis of the first few experimental values. It can be seen that the fitted and the experimental values are in good agreement for the curves shown as well as for all others i. e. in the first part of the isothermal treatment the contracion due to the loss in excess free volume is dominant. It can also be seen that the creep process (shown by circles with hyphen) is a linear function of time. A good description of the creep by a linear function is obtained for all the experimental curves where creep occurs, so this was an indication of the Newtonian type of viscous flow. The first few experimental values at higher temperatures do not fit into the logarithmic dependence possibly because of some secondary effects of the sample oscillation during its adaption to the furnace temperature. It can also be seen that for the $Cu_{60}Zr_{40}$ sample (Fig. 4) the creep decreases at the highest temperature. This is a consequence of the creation of the conditions for the nonlinear flow.

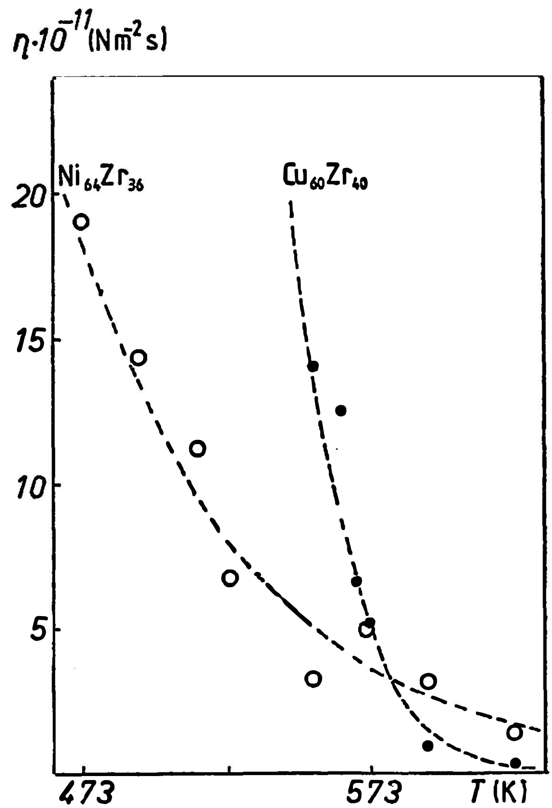

Fig. 5. The coefficient of viscosity as a function of temperature. The full points are for the $Cu_{60}Zr_{40}$ samples and the empty circles for the $Ni_{64}Zr_{36}$ samples.

The results for the coefficient of viscosity for the both investigated amorphous systems obtained by numerical analysis of the creep process are given on Fig. 5 and Table 2.

The obtained values for the coefficient of viscosity can be approximated by the functional dependence of the type

$$\eta = \eta_0 \exp (\Delta E/kT).$$

TABLE 2

	$Ni_{64}Zr_{36}$	$Cu_{60}Zr_{40}$
Temperature (K)	$\eta \times 10^{-11}$ (Pa · s)	$\eta \times 10^{-11}$ (Pa · s)
473	19.03 ± 0.004	—
493	14.06 ± 0.004	—
513	11.96 ± 0.004	—
523	6.71 ± 0.004	—
553	3.25 ± 0.004	13.98 ± 0.004
563	—	12.51 ± 0,004
568	—	6.55 ± 0.004
573	5.13 ± 0.004	5.26 ± 0.004
593	3.14 ± 0.004	0.92 ± 0.004
623	1.39 ± 0,004	0.28 ± 0.004

The coefficient of agreement was for the $Ni_{64}Zr_{36}$ system 0.92 and for the $Cu_{60}Zr_{40}$ system 0.98. Such a good agreement with the above mentioned functional dependence enabled us to evaluate the activation energies for the creep process, because the value $\Delta E/kT$ can be directly obtained by fitting. For the $Ni_{64}Zr_{36}$ system the evaluated activation energy was 41.1 kJ/mole and for the $Cu_{60}Zr_{40}$ system 78.8 kJ/mole. A great difference in the activation energies could be expected because of the phase diagram of the both systems.

The obtained activation energies as well as the behaviour of the viscosity can be explained on base of the following results[4-6]:

i) in comparison with the corresponding Cu-Zr alloys, there is a stronger interaction between the neighbours in the Ni-Zr alloys;

ii) the Au-Zr system can be described by the model of dense random packed hard spheres;

iii) a local ordering during melting of the Ni-Zr system was noticed; the melting point coincides with the melting point of the ideal solution.

During the formation of the Cu-Zr system there will be quenched in a great amount of the free volume excess. The consequence is that creep occurs sufficiently later in this system than in the Ni-Zr system. The atom migration in the Cu-Zr system will lower the free volume and its disappearance will create the conditions for creep. The sample in this conditions will flow very intensively because of the weak interaction between the atoms. This explanation can be used for the interpretation of the nonlinear contribution of the creep in Fig. 4.

In the Ni-Zr system, where there is not much of the quenched in free volume, the creep process occurs much earlier. The contraction of this sample is sufficiently smaller than the contraction of the Cu-Zr sample.

3. *Conclusion*

We have investigated the temperature dependence of the coefficient of viscosity for $Cu_{60}Zr_{40}$ and $Ni_{64}Zr_{36}$ amorphous systems. The investigations were performed under applied tensile stress in the temperature range from 473 K to

623 K. It was found out that $Ni_{64}Zr_{36}$ system has a significant lower activation energy for creep than the $Cu_{60}Zr_{40}$ system, so the creep influenced by the applied tensile stress was not observed at temperatures lower than 553 K. Up to this value the process of contraction due to the loss in excess free volume was dominant. Although there was a significant difference in the numerical values of the coefficient of viscosity, it can be said that both systems have an identical behaviour. A great similarity can be found in the gradual domination of the creep process, which is in both cases of the isoconfigurational type.

Acknowledgments

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TEMPERATURNA ZAVISNOST KOEFICIJENTA VISKOZNOSTI METALNIH STAKALA $Cu_{60}Zr_{40}$ i $Ni_{64}Zr_{36}$

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Vlastitom metodom hladnog prsta izvršeno je dilatometrijsko mjerenje amorfnih sistema $Cu_{60}Zr_{40}$ i $Ni_{64}Zr_{36}$ pod uticajem napona pri povišenim temperaturama. Separiranjem procesa napuštanja slobodnog volumena iz eksperimentalnih podataka dobiven je odgovor na pitanje da li uzorak teče. Uočeno je da se ovaj proces javlja na nižim temperaturama za sistem $Ni_{64}Zr_{36}$ i da ima linearnu vremensku zavisnost. Izvršeno je računanje koeficijenta viskoznosti na osnovu dobivenih podataka o tečenju uzoraka u oba posmatrana sistema, data njihova funkcionalna ovisnost o temperaturi i na osnovu nje određena aktivaciona energija za oba sistema. Zavisnost koeficijenta viskoznosti o temperaturi može se prikazati kao $\eta = \eta_0 (\Delta E/kT)$. Dobivena aktivaciona energija za sistem $Ni_{64}Zr_{36}$ iznosi 41.1 kJ/mol, a za sistem $Cu_{60}Zr_{40}$ njena vrijednost je 178.8 kJ/mol.