Kinematic viscosity of liquid Al-Cu alloys

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N Yu Konstantinova and P S Popel
Ural State Pedagogical University, 26 Cosmonavtov Ave., Ekaterinburg 620017, Russia
E-mail: konstantinova@k66.ru

Abstract. Temperature dependences of kinematic viscosity \( \eta \) of liquid Al\(_{100-x}\)-Cu\(_x\) alloys (\( x = 0.0, 10.0, 17.1, 25.0, 32.2, 40.0 \) and 50.0 at.\%) were measured. A technique based on registration of the period and the decrement of damping of rotating oscillations of a cylindrical crucible with a melt was used. Viscosity was calculated in low viscous liquids approximation. Measurements were carried out in vacuum in crucibles of BeO with a temperature step of 30\(^\circ\)C and isothermal expositions of 10 to 15 minutes during both heating up to 1100-1250\(^\circ\)C and subsequent cooling. We have discovered branching of heating and cooling curves \( n(T) \) (hysteresis of viscosity) below temperatures depending on the copper content: 950\(^\circ\)C at 10 and 17.1 at.\% Cu, 1050\(^\circ\)C at 25 and 40 at.\% Cu, 850\(^\circ\)C at 32.2 at.\% Cu. For samples with 10 and 17.1 at.\% Cu the cooling curve "returns" to the heating one near 700\(^\circ\)C. An abnormally high spreading of results at repeated decrement measurements was fixed at heating of the alloy containing 50 at.\% Cu above 1000\(^\circ\)C. During subsequent cooling the effect disappeared. Isotherms of kinematic viscosity have been fitted for several temperatures.

1. Introduction
Aluminum-copper alloys are widely used in various fields of engineering. Their physical properties (electrical and thermal conductivity, ductility, resistance to corrosion, etc.) are determined by the concentration proportions of the initial components. The technological processes of production of aluminum-copper alloys are connected with the transformation of the initial materials into a melted state and their subsequent crystallization. The results of numerous investigations show that most aluminum alloys retain a microheterogeneous state for a long time after melting. It may be irreversibly destroyed as a result of a superheat over the liquidus up to the temperatures characteristic of each the systems [1]. Such a superheat causes considerable changes in crystallization kinetics and, as a result, modify the crystalline structure that is formed in the course of solidification. Information on the temperatures of this transition may be obtained as a result of measuring temperature dependences of different melt properties in the course of heating after melting and subsequent cooling. In this case the indicator of the transition is the branching of the heating and cooling curves (hysteresis of properties), and the branching point is assumed to be the temperature of the transition. One of the properties most sensitive to the process of the melt homogenization is its kinematic viscosity \( \eta \).

Previously, in the regime of heating and subsequent cooling, measurements of the kinematic viscosity were made only for aluminum-copper alloys with a low content of copper (0.43; 0.85; 1.7; 2.6; 3.56; 5.48; 9.60; 16.4 and 22.1 at.\% Cu) [2]. A wider range of compositions for the aluminum-copper system (72; 76; 79; 83; 87; 91; 95 at.\% Cu) was investigated in [3], but only for two temperatures: 1100\(^\circ\)C and 1200\(^\circ\)C.
Our paper presents the results of measuring the kinematic viscosity of melts of the aluminum-copper system in the regime of heating and subsequent cooling at copper concentration up to 50 at.% in the temperature range from the liquidus temperature to 1100-1250°C.

2. Experimental

For measuring the kinematic viscosity method based on fixation of the period and the damping decrement of rotating oscillation of a cylindrical crucible with a liquid metal was used. The schematic diagram of the working part of the viscosimeter on which measurements were conducted is given in reference [1]. The values of the kinematic viscosity were calculated by the following formula:

\[ \nu = \frac{1}{\pi} \left( \frac{K}{MR} \right)^2 \left[ \delta - \left( \delta_0 / \tau_0 \right) \right] \frac{2}{\tau \sigma^2}, \]  

where \( M \) and \( R \) are the mass of melt and the radius of the crucible; \( K \) is the moment of inertia of a suspending system; \( \delta, \delta_0, \tau, \tau_0 \) are damping decrements and vibration periods of the system with and without a melt, respectively; \( \sigma \) is the quantity which takes into account the effect of the sample butt-end surfaces on the vibrations. This calculation formula was derived by E.G. Shvidkovsky as a result of solution of the inner hydrodynamic problem for low-viscous liquids, among which are melted metals and most of alloys [4].

To increase the precision of measurements of the damping decrement, we developed a special system of registration of the amplitude of rotating oscillation. Vibration of a light beam reflected from the mirror of a suspending system onto a transparent ruler recorded by a video camera, and then, after computer processing of the video image, a plot of the amplitude of damped vibrations versus the vibration number was built using standard computer software. This improved considerably the reliability of determination of the damping decrement, which resulted in an increase in the precision of measurements of viscosity from 5% to 1-2%.

To increase the viscosimeter sensitivity, the parameters of its suspending system were optimized: use was made of crucibles of beryllium oxide with an inner radius of 8.4 mm. The sample mass was near 30 g, the moment of inertia of the suspending system was equal to \( 8.58 \times 10^{-6} \text{ kg} \cdot \text{m}^2 \), the diameter of the Ni-Cr alloy filament was 0.24 mm, and the period of vibrations was near 3.5 s. The above-mentioned parameters of the suspending system ensured the feasibility of the conditions for which the formula (1) was obtained:

\[ H \geq 1.85 R, \]  

\[ y = \frac{2\pi R^2}{\tau \nu} \geq 100, \]

where \( H \) is the sample height; \( R \) is its radius; \( \tau \) is the period of vibration; \( \nu \) is the kinematic viscosity.

Investigation were made of samples of aluminum-copper alloys at copper concentrations of 0.0, 10.0, 17.1, 25.0, 32.2, 40.0 and 50.0 at.%. In choosing them we proceeded from the phase diagram of the aluminum-copper system presented in [5].

Before the beginning of an experiment the sample was subjected to vacuum processing at 1100°C with the aim of removing the oxide film from its surface. Then it was cooled at a rate of 600°C/hour to room temperature. In the course of subsequent heating temperature dependences of the kinematic viscosity were taken in temperature intervals from the liquidus line to 1100°C and back. A sample which contained 50 at.% Cu was heated up to 1250°C. Measurements were conducted in vacuum with a temperature step of 30°C and isothermal exposition of 10-15 min at every point.

3. Results

The temperature dependences of the kinematic viscosity obtained by the authors for the investigated alloys are presented in figure 1.
Figure 1. Temperature dependences of kinematic viscosity of liquid Al-Cu alloys. Accuracy of the measurements for all the compositions except heating curve of the Al-50 at.% Cu alloy is shown as the arrow in the left down corner.

In their analysis attention is attracted by the branching of the curves $\nu(T)$ obtained in the course of heating and subsequent cooling (viscosity hysteresis). The branching temperature depend on the copper concentration and are 900°C for pure aluminum, 950°C for alloys containing 10 and 17.1 at.% Cu, 1050°C at 25 and 40 at.% Cu and 850°C at 32.2 at.% Cu. In the course of heating of the alloy containing 50 at.% Cu, beginning with 1000°C, abnormally high spread in values of the damping decrement was recorded in repeated measurements. It complicated the determination of the branching point – it was only clear it was above 1120°C – and disappeared during subsequent cooling. For liquid aluminum and samples that contain 10 and 17.1 at.% Cu one can observe “a return” of the curves $\nu(T)$ obtained in cooling to the corresponding heating curves near 700°C. Previously a similar behavior of temperature dependences of viscosity for liquid aluminum and hypoeutectic Al-Cu alloys was observed by Zamyatin et al. [2,6]. On the whole it may be noted that in most cases temperature dependences of viscosity are “smoother” in cooling than in preceding heating.

Isotherms of kinematic viscosity for several temperatures built by the data obtained while cooling the samples are shown in figure 2. Attention is drawn to clearly defined maximum and minimum of viscosity close to the stoichiometric compositions of Al$_3$Cu and Al$_2$Cu, correspondingly. After the last extremum a tendency to following viscosity growth is rather distinct too.

In frames of activation models of a liquid flow, the temperature dependence of viscosity is usually described by the exponential function. Corresponding $\ln \nu(T^{-1})$ fit is a straight line. Its slope defines the activation energy of the flow $E_a$. We have calculated the concentration dependence of the activation energy and fitted it in figure 3. Attention is attracted to rather complicated form of the $E_a(x_{Cu})$ curve with a non-monotonous fragment at compositions corresponding to above mentioned extremums and following increase to equiatomic concentration.
4. Discussion

It was mentioned above that branching of $v$ vs. temperature curves corresponding to heating the sample after its melting and to its subsequent cooling are usually interpreted as a sign of irreversible structural transformation in a melt. We think that the most probable nature of this transformation is irreversible transition the melt from a metastable microheterogeneous state inherited from crystalline precursors to a state of true solution.

Nevertheless, a different kind of viscosity hysteresis in hypo- and hyper-eutectic melts is obvious. We link it preliminary with the specific reversible short-range order rearrangement in liquid aluminum and hypo-eutectic Al-Cu melts discovered in viscometric experiments of Zamjatin et al. [2] and confirmed by results of X-ray diffraction study of Bazin et al [6].

Growth of the viscosity and the activation energy of viscous flow near 50 at.% Cu can be connected with strengthening of interaction of different atoms while closing the melt composition to $\text{AlCu}_2$ where some peculiarities on isotherms of properties were fixed systematically.

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