PROCEEDINGS OF THE IV INTERNATIONAL CONFERENCE/HIGH TEMPERATURE CAPILLARITY

JOURNAL OF MATERIALS SCIENCE **40** (2005) 2217–2219

Surface tension of melts of Na-K-Cs ternary system

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In this work the results of the experimental study of surface tension (ST) of alkali metals and their Na-K-Cs ternary system over the whole composition range and for temperatures from the melting point to 680 K are presented. Careful thermal treatment for several hours at 700 K under 10^{-7} Pa vacuum condition and a new way of loading the measuring cell have been used to exclude possible contamination of the liquid metals and the measuring cell. To measure ST the sessile-drop method, which yields an experimental error of 1% with 95% probability, was used. Contents of impurities were not larger than 0.005%.

Surface tensions of 110 Na-Cs-K alloys have been measured over the whole composition triangles for the first time. Polytherms of ST for pure metals and ternary alloys are well described by linear equations. The isotherms of ST for each Na/Cs + K section have been constructed. It was found that K is a surface-active addition in ternary alloys when $X_{Na}:X_{Cs} > 14:1$, while it becomes a surface-inactive component if $X_{Na}:X_{Cs} < 14:1$. All ternary alloys along the $X_{Na}:X_{Cs} = 14:1$ section have the same ST, independent from K content. Lines of equal values of ST have been plotted for the whole composition triangles as well.

1. Introduction

Alkali metals and their alloys, having a unique combination of such properties as extremely high values of thermal and electric conductivities, the lowest melting temperatures (up to −80°C for some alloys), small values of ionization potential, electronic work function, specific weight and viscosity, high values of surface tension and critical temperature, and so on, attract the investigators' attention. They are widely used in nuclear reactors, new power-intensive chemical current sources, emission electronics, transformers of thermal energy into electric energy, airspace materials, medicine, and so forth. Multicomponent liquid metallic coolants based on alkali metals are a new type of coolants in nuclear reactors [1–3].

There are many works in literature devoted to the investigation of physicochemical properties of alkali metals and their alloys. However, the surface properties of the latter are not studied well enough due to their high chemical activity [4, 5]. In this work we present the new results of experimental measurements of the ST of pure alkali metals and their ternary alloys.

2. Device and technique for the study of the surface tension of liquid alkali metals and their multicomponent alloys

For the surface tension measurements the well-known sessile-drop method was used [6]. It is the most precise method. We elaborated a new combined device [7], which allows to form alloys in the entire concentration range and to measure their ST and electronic work function (EWF), all in a single loading. This device permits forming alloys based on either component A or B, of desired concentration and at any stage of the experiments, all without opening the measuring cell. The device enables one to measure the ST and EWF of an alloy in identical experimental conditions. It does not allow a contact with the external environment, and enables preparation of a large number of alloys with necessary compositions without unsealing the apparatus. It also allows measuring the ST and EWF for the same alloy many times, renovating the surface of a drop every time. For this technique the density of the alloys must be measured separately. The improved two-capillary pycnometer was used for this purpose. The use of the combined device decreases the duration of measurements by a factor of ten, and yields an essential economy of metals. The measurement errors were 0.5% for the ST, 0.1% for the density, and 1% for the electronic work function.

3. Surface tension of alkali metals and their ternary alloys

Using the combined device and specially elaborated techniques we measured the ST of pure alkali metals and the alloys of the Na-K-Cs system. The high purity alkali metals with impurity content of no more than 0.005% for Na and Rb and 0.12% for Li were used. The measurements were made over the temperature range 333 to 623 K while both increasing and decreasing the temperature.

No data on the ST of ternary alloys of alkali metals is available in the literature other than experimental results on a density for 11.8 at.% of Na + 47.4 at.% of

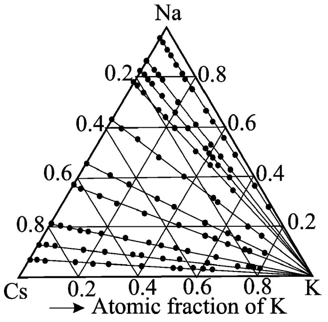

Figure 1 Compositions of 110 ternary alloys of the Na-K-Cs system along the sections, for which the surface tension was measured.

$K + 40.8$ at% of Cs alloy in the temperature range from 199 to 300 K [8], and a phase diagram for a single Na-K-Cs system [9]. Beside the combination of the unique properties of pure alkali metals, the ternary alloys have other valuable properties, for example, a wide temperature range of the liquid state. For the Na-K-Cs system it extends from 195 to 2000 K (the critical temperature is $T_c \sim 3000$ K). This property is of great importance due to its application in nuclear reactors. The experimental results on the ST of 110 alloys of the Na-Cs-K system are presented in this paper for the first time.

The alloys of the Na-K-Cs system, formed along ten sections, enclose the entire composition triangle. The alloys were prepared by adding K to a binary Na-Cs alloy at a constant X_{Na}:X_{Cs} ratio. Here X_{Na} and X_{Cs} are concentrations of Na and Cs. (Fig. 1).

4. Discussion

The polytherms of ST of pure metals over the investigated temperature ranges are described by linear equations:

$$\sigma = \sigma_m - a(T - T_m), \qquad (1)$$

where T_m is the melting temperature, σ_m is the ST at T_m, and a is the temperature coefficient of ST. Values of T_m, σ_m and a for pure alkali metals are given in Table I. The results calculated with Equation 1 may be recommended as the most reliable.

The polytherms of ST of the ternary alloys are described by linear equations with negative or positive temperature coefficients, depending on the concentration of K. The polytherm of the ST of the ternary eutectic alloy is given by equation

$$\sigma(T) = 81.38 - 0.024(T - T_e), \qquad (2)$$

TABLE I Values of T_m, σ_m and a in Equation 1 for pure alkali metals

Metal	Li	Na	K	Rb	Cs
T_m (K)	453.4	370.86	336.71	312.50	301.40
σ_m (mN/m)	419	205.3	116.2	95.8	75.0
$-a$ (mN/m · K)	0.150	0.094	0.062	0.052	0.050
Temp.range (K)	454–700	371–523	337–490	313–435	302–423
δ^a(%)	0.8	0.4	0.4	0.5	0.5

[a] δ—the root-mean-square deviation of the experimental results from Equation 1.

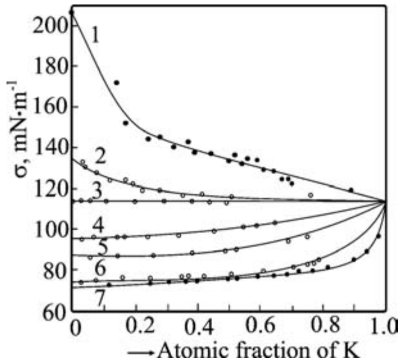

Figure 2 Isotherms of a surface tension of the ternary alloys of the Na-Cs-K system at 373 K along the sections with X_{Na}:X_{Cs} = const: 1—Na-K; 2—X_{Na}:X_{Cs} = 58:1; 3—14:1; 4—6:1; 5—2:1; 6—1:2; 7—Cs-K

where $T_e = 195$ K is the temperature of the ternary eutectic alloy. The root-mean-square (rms) deviation of the experimental results from that by Equation 2 is about 0.6%.

The isotherms of the ST of ternary alloys at 373 K along five sections with constant X_{Na}:X_{Cs} ratio are shown in Fig. 2. It was found that K is a surface-active component in the Na-K system (Fig. 2, curve 1), while it was a surface-inactive component in the Cs-K system (Fig. 2, curve 7). Peculiarity of the influence of K on the ST of Na-Cs-K alloys is in variation of its surface activity when Na is replaced by Cs in the initial Na-Cs alloy. As it is shown in Fig. 2, K is a surface-active additive in ternary alloys when X_{Na}:$X_{Cs} > 14$:1 (curves 1 and 2), and it becomes a surface-inactive additive when X_{Na}:$X_{Cs} < 14$:1 (curves 4 to 7). This phenomenon may be regarded as a "concentrational inversion of a surface activity" of a component in a ternary system. Surface tensions of Cs, K and Na are 71, 113.8 and 203.1 mN/m, respectively. The isotherm 3 (Fig. 2, curve 3) shows that all ternary alloys along the section beginning with the 93.3 at.% Na + 6.7 at.% Cs binary alloy have the same surface tension of 113.9 mN/m at 373 K, independent of the content of K in the alloy. Independence of the ST of a ternary alloys from the content of one of the components at constant ratio of concentrations of the other two components is called "concentration buffering of a surface tension". The curves of equal values of ST (isolines) of the Na-Cs-K alloys at 373 K are shown in Fig. 3. Isoline 6 in Fig. 3 is a line of "concentration buffering of ST" at X_{Na}:X_{Cs} = 14:1.

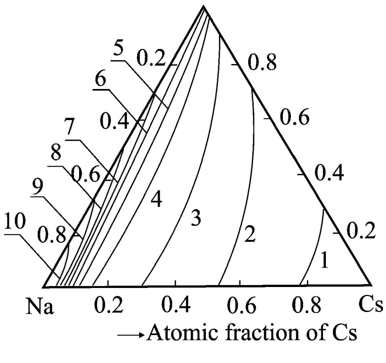

Figure 3 Isolines of a surface tension of the Na-Cs-K system at 373 K: 1—75; 2—80; 3—90; 4—100; 5—110; 6—113.8; 7—120; 8—130 mN/m.

5. Conclusion

For the first time of surface tensions of 110 Na-Cs-K ternary alloys were studied. It is shown that the polytherms of ST for pure alkali metals and their binary and ternary alloys over the investigated temperature ranges are described with linear equations. The ST isotherms appear as the smooth curves without any extreme points. One can concluded that K is a surface-active additive to Na-Cs binary alloy when X_{Na}:X_{Cs} $>$ 14:1, while it becomes a surface-inactive one when X_{Na}:X_{Cs} $<$ 14:1.

This phenomenon may be regarded as a concentrational inversion of a surface activity of K in Na-Cs-K ternary alloys.

It is found that the addition of K to initial 93.3 at.% of Na + 6.7 at.% of Cs (X_{Na}:X_{Cs} = 14:1) binary alloy does not change the ST values of the ternary alloys along the whole section. It is a concentration buffering of a surface tension of the ternary alloys.

The obtained surface tension values for pure alkali metals (Table I) may be recommended as the most reliable values.

The ternary eutectic alloy of Na-Cs-K system is liquid over the wide temperature range (from 195 to 2000 K) and therefore may be recommended as effective liquid heat-coolant in nuclear reactors.

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Received 31 March
and accepted 20 October 2004