Fluid Phase Equilibria 421 (2016) 39–48

Contents lists available at ScienceDirect

Fluid Phase Equilibria

journal homepage: www.elsevier.com/locate/fluid

Density, surface tension and viscosity of liquid binary Al-Zn and ternary Al-Li-Zn alloys

M.E. Trybula*, T. Gancarz, W. Gąsior

Institute of Metallurgy and Materials Science, Polish Academy of Sciences, Krakow, Poland

ARTICLE INFO

Article history:
Received 23 January 2016
Received in revised form
16 March 2016
Accepted 19 March 2016
Available online 23 March 2016

Keywords:
Density
Surface tension
Viscosity
Liquid Al-Zn alloys
Liquid Al-Li-Zn alloys

ABSTRACT

Three thermophysical properties: density, surface tension and viscosity of liquid binary Al-Zn and ternary Al-Li-Zn alloys have been investigated in this work, by means of the draining crucible method. Three Al-Li-Zn compositions corresponding to distinguished intermetallic phases, i.e. Al_5Li_3Zn, $Al_{35}Li_{32}Zn_{33}$ and $AlLiZn_3$, have been selected for investigation. Experimentally determined surface tension of ternaries has been compared to the modelled one, using the commonly known Butler's model and two sets of ternary interaction parameters given in the literature. Whereas, viscosity calculations for the ternary Al-Li-Zn alloys have been performed using three thermodynamics-based models, selected for their reliability, and confronted to the experimental data measured in this work. Both, measured and modelled surface tension decreased with increasing Li and Zn content, opposite to viscosities exhibiting an increase with Li and Zn content raise. Whereas measured densities of the investigated ternaries are decreasing with Li and Zn content growth in confrontation to the three respective Al-Zn binaries investigated in this work.

1. Introduction

The knowledge of phase diagram and physicochemical properties is fundamental for designing new performance materials possessing improved mechanical properties and setting the conditions of technological processes. More importantly, thermophysical properties of ternary Al-Li-Zn alloys, still unknown, is of a great importance to optimize the technological processes such as melting or casting and extracting. Al-Li based ternary alloys, i.e. Al-Li-X (where X stands for Cu, Mg, Zn), are characterized by superior mechanical properties, these are low specific weight, high strength and high elasticity modulus. It has also been observed that ternary Al-X-Zn systems (where X stands for Mg, Li) show metastable quasicrystals existence in certain concentration ranges, which have recently been the subject of impressive experimental and theoretical contributions [1–3].

Although some systems, for instance the Al-Zn alloys, are almost fully recognized, however, it is still required to update their properties. Several interesting papers concerning solid materials' examinations have been reported in the literature in confrontation to smaller ones found for liquid properties of Al-Zn alloys [4–7]. Whereas, the Al-Li based alloys have recently received considerable attention as potential lightweight replacements for Al based alloys in light metal industry for aerospace applications. For instance, the addition of 1.8–2.1 at.% Li remarkably altered the precipitation behaviour of Al-Cu-Mg-Zn-Li alloys, which are the highest strength aluminium alloys known [8].

According to the previous experimental and theoretical reports, concerning thermodynamics of liquid Al-Zn alloys, positive deviations from ideal solution were observed, therefore signifying the phase segregation tendency occurring in liquid state [6]. In relation to the previously established thermophysical properties of liquid Al-Zn alloys, measured viscosity [9] and surface tension [4] exhibit significant negative deviation from their linear concentration dependence in comparison to density changing almost linearly with increasing Zn content [9]. Whereas thermodynamic studies for the two remaining binary systems, i.e. Al-Li and Li-Zn alloys, revealed negative deviations from ideal solution which were also confirmed and profoundly described in our last paper, concerning theoretical and experimental study of bulk and surface properties of those binaries [10].

Contrary to the records for limiting binaries mentioned above, any information on density, viscosity and surface tension for liquid ternary Al-Zn-Li alloys have not yet been found in the literature. Only one property has been given, the mixing enthalpy for three ratios of constant molar fraction of Al to Zn measured and modelled

* Corresponding author.
E-mail addresses: trybulamarcela@wp.eu, m.trybula@imim.pl (M.E. Trybula).

http://dx.doi.org/10.1016/j.fluid.2016.03.013
0378-3812/

by Kim & Sommer [11]. Later, Guo et al. [12] devised the ternary interaction parameters for thermodynamic description of the liquid phase, and very recently we proposed proposed a new set of those [13], which allowed to better describe the thermodynamics of liquid Al-Zn-Li alloys including partial and mixing thermodynamic functions obtained from electromotive force measurement.

In correspondence to the solid state of the Al-Li-Zn system, it is important to mention a recent review of Bodak [14], compiling the full information on all works done for this system and containing important observations concerning intermetallic phases, i.e. $\tau(Al_5Li_3Zn)$, $\upsilon(Li_{26}Al_6(Al_{1-x}Zn_x)_{49})$ and $\gamma(AlLiZn_3)$, experimentally determined by Badeva & Sald'au [15]. Some interesting structural characteristics of these phases had been released and therefore, thermophysics knowledge of the three aforementioned Al-Li-Zn alloys' compositions is essential for their further investigation. Later Guo et al. applied this knowledge for phase diagram calculation [12].

According to previous Trybula et al. [10] and Gancarz et al. [16,17] studies, the draining crucible method, applied in this work, is a promising experimental technique enabling to determine three thermophysical properties of dense liquids at one measurement, efficient for both light and heavy metals. Apart from the scientific reports mentioned above, related to methodology and data found, the proposition of this work is to show the capability of the draining crucible method to correctly measure the three physicochemical properties of the Al-Zn binary alloys. We chose Al-Zn alloys because complete thermophysical data existed in the literature for them and, therefore, it builds a correct background for further analysis of these properties in the liquid Al-Li-Zn alloys.

The primary aim of this work is to examine the influence of Li addition on density, viscosity and surface tension change measured in the 823–1023 K temperature range for three liquid binary Al-Zn alloys. Three different ratios of constant Al to Zn molar fraction have been taken into consideration which correspond to the compositions of three intermetallic phases mentioned above, occurring in the solid state of Al-Li-Zn alloys. In the first stage, three thermophysical properties of liquid Al-Zn alloys are discussed with regard to the available literature data. Further, these properties for liquid Al-Li-Zn alloys are determined and discussed with modelled data obtained in this work, using two assumptions in order to verify the existence of a ternary effect, which could correspond to an associative tendency previously observed in liquid Al-Li and Li-Zn alloys [10]. Finally, this work is addressed to build a complete database of thermophysical properties, including the three limiting binary alloys and Al-Li-Zn alloys, examined with use of the draining crucible (DC) method. The viscosities of Al-Zn-Li alloys are computed using the most representative thermodynamics-based models, as mentioned and used in Ref. [10], whereas, Butler's model [18] was employed to expand the discussion of experimental surface tension values.

2. Methodology

2.1. Experiment

Binary and ternary Al-Zn based alloys were prepared using high purity Al, Li and Zn metals (see details in Table 1), further melted in a molybdenum crucible which was placed in a furnace. To provide the best possible protective atmosphere, every experiment was conducted in a glove box filled with high purity argon. The level of oxygen, water vapour and nitrogen were kept constant and it was below 1 ppm as monitored by a solid-state analyser. Generally, three binaries: $Al_{75}Zn_{25}$, $Al_{51}Zn_{49}$, $Al_{83}Zn_{17}$ and ternaries: $\tau(Al_5Li_3Zn)$, $\upsilon(Al_{33}Li_{32}Zn_{33})$, $\gamma(AlLiZn_3)$ were the subject of presented and discussed herein experimental and theoretical

Table 1
Chemical compositions[a] of Al-Zn binaries and Al-Li-Zn ternaries considered in this work.

Metals	Fraction purity		
Al, Zn – rods*	0.99999		
Li – rods*	0.99950		
Chemical formulae	Al-Zn alloys		
	X_{Al}[b]		X_{Zn}[b]
$Al_{83}Zn_{17}$	0.83		0.17
$Al_{51}Zn_{49}$	0.51		0.49
$Al_{25}Zn_{75}$	0.25		0.75
	Al-Li-Zn alloys		
	X_{Al}[b]	X_{Li}[b]	X_{Zn}[b]
Al_5Li_3Zn[#]	0.56	0.33	0.11
$Al_{28}Li_{26}Zn_{27}$[#]	0.35	0.32	0.33
$AlLiZn_3$[#]	0.20	0.20	0.60

[a,b] Standard uncertainties u are u(X) = 0.005 for Al and Zn, whereas u(X) = 0.01 for Li.
*Sigma Aldrich Distributor.
[#] Al-Li-Zn alloy compositions and corresponding them signature used in this work: $\tau(Al_5Li_3Zn)$, $\upsilon(Al_{28}Li_{26}Zn_{27})$, $\gamma(AlLiZn_3)$.

contributions, which are plotted in Fig. 1, inscribed in a Gibbs triangle.

Appropriately prepared samples were subjected to simultaneous measurement of three thermophysical properties: density, surface tension and viscosity using the draining crucible (DC) method. In this work, we give only basic formulae which are used, in practise, for determination of the aforementioned properties, however, a reader interested in details of the DC method is referred to Refs. [10,19–21].

Generally, a liquid alloy is flowing out through a hole at the bottom of a crucible under gravity and the cumulative weight of the liquid as a function of time is measured, and described by a polynomial:

$$A_m = \sum_{i=1}^{n} A_i t^{i-1}, \tag{1}$$

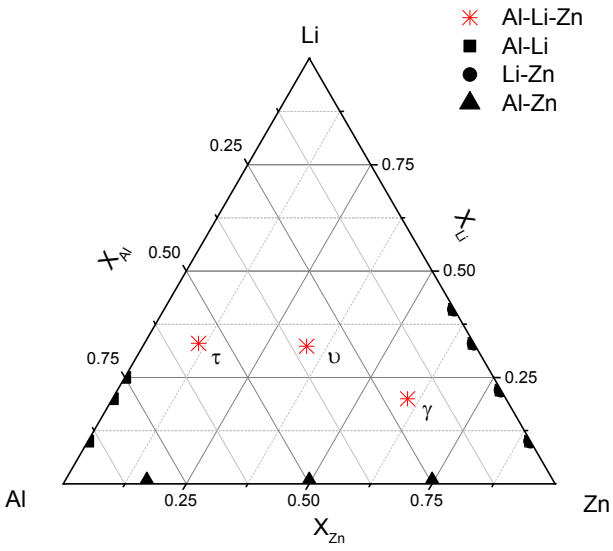

Fig. 1. τ, υ and γ ternary Al-Li-Zn alloys depicted in Gibbs triangle (chemical compositions, see Table 1). ■, ● and ▲ closed triangle represent three reference binary systems, Al-Li, Li-Zn and Al-Zn, respectively.