Surface tension of binary and ternary aluminium alloys of the systems Al–Si–Mg and Al–Zn–Mg

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The surface tension and density of liquid binary and ternary aluminium alloys of the systems Al–Si–Mg and Al–Zn–Mg (Si, Mg and Zn contents less than 19, 8 and 20 wt %, respectively) have been measured by means of the maximum bubble pressure method. A semi-empirical theory, which relates the surface tension to bulk thermodynamic properties, is used to calculate the surface tension of the binary alloys and discuss the experimental data. For the ternary alloys, the present results indicate that in the range of compositions explored here, the properties of the ternaries can be obtained from those of the binaries. Comparison with results previously reported by other authors is made.

1. Introduction
The surface tension of liquid metals [1, 2] is both a fundamental property and a parameter of great technological importance. In particular, the surface tension of liquid aluminium and its alloys is an essential input in developing any casting model [3, 4] and in understanding the wettability at the aluminium/ceramic system, a property of crucial relevance in the fabrication of metal matrix composites [5]. Despite its importance, the amount of work addressed to measure the surface tension of commercial and ternary alloys [4], is much less than that dedicated to the binary systems [6–15].

In this work we present results for the surface tension, σ, and density, ρ, of aluminium alloys of the ternary systems Al–Si–Mg and Al–Zn–Mg obtained by means of the maximum bubble pressure (MBP) method. Partial accounts of the results discussed here were reported previously [14, 15]. The experimental data will be used to elucidate whether σ and ρ of the ternary alloys can be obtained from data for the binary systems [15]; this question is of both practical and fundamental interest. A semi-empirical theory, which relates the surface tension to bulk thermodynamic properties [16–18], will be used to calculate the surface tension of the binary alloys and discuss the experimental data.

As a final remark, we should point out that all data presented here for the surface tension of the liquid alloys should be associated with the oxidized surface, in the sense of [19]. This assessment is supported by recent findings [20] which indicate that the changes in the maximum pressure observed previously [14, 15] that were related to changes in surface tension, do in fact correspond to modifications in the wetting conditions at the metal–capillary tip interface. The conclusion drawn previously [20] is that the MBP method does not allow us to establish a clear relation between surface tension and the degree of oxidation, and that the only reliable values are those for the oxidized surface.

2. Experimental procedure
As a detailed description of the MBP experimental apparatus utilized in this work can be found elsewhere [14], here we shall only mention some of the main features of the present experiments. Alumina Degussit Al23 capillaries with external diameter around 2 mm and wall thickness ~ 0.25 mm were used; actual diameters were measured with an accuracy of ± 0.001 mm by means of high-precision gauge techniques. The immersion depth was varied in the range 15–30 mm, whereas the bubbling rate was fixed at around 10–20 bubbles/min. To speed up the stabilization of the maximum pressure, argon N55 plus 100 p.p.m. oxygen was used as bubbling gas (see [21]). The immersion depth was measured by means of a high-precision micrometre to within ± 0.005 mm; a sudden pressure rise allowed determination of the zero point with an error of ± 0.01 mm.

The alloys investigated in this work were prepared in alumina crucibles of inner diameter around 50 mm,
TABLE I Density, \( \rho \), and surface tension, \( \sigma \) as measured in this work \((T = 973 \text{ K})\), for Al-Si, Al-Zn and Al-Mg alloys.

<table>
<thead>
<tr>
<th>Si (wt %)</th>
<th>( \rho ) (g cm(^{-3}))</th>
<th>( \sigma ) (mJ m(^{-2}))</th>
<th>Zn (wt %)</th>
<th>( \rho ) (g cm(^{-3}))</th>
<th>( \sigma ) (mJ m(^{-2}))</th>
<th>Mg (wt %)</th>
<th>( \rho ) (g cm(^{-3}))</th>
<th>( \sigma ) (mJ m(^{-2}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.38</td>
<td>866</td>
<td>2.5</td>
<td>2.42</td>
<td>865</td>
<td>0.8</td>
<td>2.37</td>
<td>856</td>
</tr>
<tr>
<td>3</td>
<td>2.39</td>
<td>865</td>
<td>5.8</td>
<td>2.46</td>
<td>862</td>
<td>3</td>
<td>2.34</td>
<td>822</td>
</tr>
<tr>
<td>5</td>
<td>2.40</td>
<td>858</td>
<td>9.8</td>
<td>2.52</td>
<td>857</td>
<td>5</td>
<td>2.33</td>
<td>798</td>
</tr>
<tr>
<td>8</td>
<td>2.41</td>
<td>855</td>
<td>19.2</td>
<td>2.71</td>
<td>846</td>
<td>8</td>
<td>2.31</td>
<td>781</td>
</tr>
<tr>
<td>12</td>
<td>2.43</td>
<td>847</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19</td>
<td>2.46</td>
<td>838</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

TABLE II Slopes of the straight lines, or at the origin of the curves (*), fitted to the experimental and theoretical results for the surface tension of the aluminium binary alloys considered in this work, as a function of the element content. The data corresponding to [6, 7] were derived from the figures reported therein.

<table>
<thead>
<tr>
<th>Experimental values (mJ m(^{-2}))</th>
<th>Theory (mJ m(^{-2}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>This work</td>
<td>[9, 10]</td>
</tr>
<tr>
<td>Si</td>
<td>-1.6</td>
</tr>
<tr>
<td>Zn</td>
<td>-1.2</td>
</tr>
<tr>
<td>Mg</td>
<td>-22.0*</td>
</tr>
</tbody>
</table>

The surface tension was calculated from a relation derived by Cantor [21] and modified by Schrödinger [22], assuming that the bubbles had spherical shape, slightly deformed due to the metallostatic pressure, and detached with a diameter that coincided with the outer diameter of the capillary (2r). Only the first correction term [21, 22] was found to be important in the present measurements; in such a case the surface tension is given by

\[
\sigma = \rho P \left(1 - \frac{2}{3} \frac{\rho g}{P_m} \right)
\]

where \( \rho \) is the density of the liquid alloy, \( g \) the acceleration of gravity and \( P \) is given by,

\[
P = P_m - \rho gh
\]

where \( P_m \) is the maximum (measured) pressure and \( h \) the immersion depth.

The method also allows the measurement of the density, \( \rho \), of the liquid metal by varying the immersion depth

\[
\rho = \frac{\Delta P}{g \Delta h}
\]

where \( \Delta P \) is the pressure change due to an increment \( \Delta h \) in the immersion depth.

The errors for the surface tension and density were estimated to be \( \pm 6 \text{ mJ m}^{-2} \) and \( \pm 0.04 \text{ g cm}^{-3} \), respectively. We finally note that, although the error in determining the density by means of the MBP is relatively high, essentially due to the high \( \sigma / \rho \) ratio of aluminium, the results (see below and [14, 15]) are in good agreement with those obtained by other authors.

3. Binary alloys

3.1. Experimental results

The results for the binary alloys investigated in this work are summarized in Tables I and II and Figs 1–3.