



Thermodynamic modelling and assessment of the Au–In–Sn system

G. Cacciamani^{a,b,*}, G. Borzone^{a,b}, A. Watson^c

^a DCCI-University of Genova, Genova, Italy

^b INSTM, Genova, Italy

^c IMR/SPEME, University of Leeds, UK

ARTICLE INFO

Article history:

Received 27 June 2008

Received in revised form

15 September 2008

Accepted 18 September 2008

Available online 21 October 2008

Dedicated to the memory of Prof. Riccardo Ferro

Keywords:

Calphad

Lead-free solders

Au–In–Sn

Thermodynamics

Alloys

ABSTRACT

As part of COST Action 531, a thermodynamic assessment of the Au–In–Sn has been carried out using experimental phase equilibrium data from the literature, along with more recent and more extensive experimental results from phase equilibrium and thermodynamic studies. As well as reproducing the experimental data very well, the self-consistent thermodynamic description produced for this ternary system is consistent with the COST 531 thermodynamic database for lead-free solders. Assessed thermodynamic parameters for the system are listed, along with crystallographic details of all stable phases.

© 2008 Elsevier Ltd. All rights reserved.

1. Introduction

The health aspects of lead in materials have been of concern for a number of years. It has been widely reported that accumulations of lead in the human body can lead to delays in neurological and physical development and disorders in the nervous and reproductive systems. Consequently, there has been a drive to reduce the amount of lead in use, and particular in relation to lead-containing solder materials. Research programmes dedicated to find lead-free alternatives to traditional lead–tin solders have been initiated world-wide, and in particular in Europe through COST Action 531 on ‘Lead-free Solder Materials’. It is now widely accepted that there is no single ideal drop-in replacement for lead–tin solder, suggesting that a number of different alloys are necessary to satisfy the numerous and diverse requirements of the electronics industry.

The aims of COST 531 were to study the chemical and physical properties of alloy systems relevant to lead-free solder materials and their interactions with substrates, producing scientific knowledge that can act as the basis of future work on the design of new

lead-free solder materials. A large part of the research involved studies of phase equilibria and thermodynamic properties of candidate alloy systems leading to the creation of an 11 component thermodynamic database for lead-free solders and substrates [1]. Among the 11 components chosen for the database were Au, In and Sn. Relatively little is known of the ternary Au–In–Sn system (see below) and hence it became of particular interest to COST 531, resulting in a programme of experimental study (phase equilibria and thermodynamics) leading to a self-consistent thermodynamic description that has since been incorporated into the COST 531 database. This paper, reporting the thermodynamic assessment of the Au–In–Sn system is part of a trilogy describing this work.

2. Literature overview

The phases of the Au–In–Sn system are listed in Table 1. In the same table, for each phase, crystal structure data, the temperature and composition ranges of stability reported in the literature are summarised. Thermodynamic models adopted in the present work are also reported: they will be introduced later.

An overview of the binary Au–In, Au–Sn and In–Sn thermodynamic assessments already appearing in the literature will be presented briefly and discussed. A thermodynamic calculation of the Au–In–Sn phase equilibria has also been published quite recently: it will also be presented and discussed in this section.

* Corresponding address: Dip. di Chimica e Chimica Industriale, Università di Genova, via Dodecaneso, 31, I-16146 Genova, Italy. Tel.: +39 010 353 6130; fax: +39 010 362 5051.

E-mail address: cacciamani@chimica.unige.it (G. Cacciamani).

Table 1

Au–In–Sn solid phases: crystal structure data, temperature and composition ranges of stability, sublattice models used in this work.

Phase name	Pearson symbol-prototype space group	Lattice parameters (nm)	Temperature range (°C)	Composition range and comments	Sublattice model
Au	<i>cF4</i> -Cu <i>Fm-3m</i>	Au: $a = 0.4072$ Au-10 at.% In: $a = 0.41060$ Au-6 at.% Sn: $a = 0.4103$	Au: <1064.33	Au–In: 0–13.3 at.% In Au–Sn: 0–6.6 at.% Sn	(Au, In, Sn)
In	<i>tI2</i> -In <i>I4/mmm</i>	In: $a = 0.3248$ $c = 0.4948$	In: <156.75	In–Sn: 0–12 at.% Sn	(In, Sn)
β -Sn	<i>tI4</i> - β -Sn <i>I41/ amd</i>	Sn: $a = 0.58308$ $c = 0.31810$ In-95 at.% Sn: $a = 0.5828$ $c = 0.3183$	Sn: 232.08–13	Au–Sn: 99.8–100 at.% Sn In–Sn: 90(?)–100 at.% Sn	(In, Sn)
α -Sn	<i>cF8</i> -diamond <i>Fd-3 m</i>	Sn: $a = 0.64892$	Sn: <13	Au–Sn: 99.994–100 at.% Sn In–Sn: 98(?)–100 at.% Sn	(Sn)
dhcp	<i>hP4</i> - α -La <i>P6₃/mmc</i> (also identified as <i>hP16</i> -Ni ₃ Ti)	–	Au–In: 641.8–180 Au–Sn: <532	Au–In: 9.6–14.5 at.% In; also denoted as α_1 Au–Sn: 8.2–9.1 at.% Sn; also denoted as β Au–In–Sn: complete substitution between In and Sn	(Au, In, Sn)
hcp	<i>hP2</i> -Mg <i>P6₃/mmc</i>	Au-20 at.% In: $a = 0.29157$ $c = 0.47930$ Au-11 at.% Sn: $a = 0.29081$ $c = 0.47859$	Au–In: <~640 Au–Sn: <521	Au–In: 12.2–22.4 at.% In; also denoted as ζ Au–Sn: 9.1–17.6 at.% Sn; also denoted as ζ Au–In–Sn: complete substitution between In and Sn	(Au, In, Sn)
Au ₄ In HT	<i>hP*</i> /o**	–	Au–In: 337–275	Au–In: 21.5–22.2 at.% In; also denoted as β	(Au) _{0.785} (In) _{0.215}
Au ₄ In LT	<i>hP26</i> -Cu ₁₀ Sb ₃ <i>P-3</i>	$a = 1.0545$ $c = 0.4769$	Au–In: <~280	Au–In: 21.7–22.2 at.% In; also denoted as β_1 or Au ₇ In ₂	(Au) _{0.778} (In) _{0.222}
Au ₃ In HT	<i>o**</i>	–	Au–In: 487–339.5	Au–In: 24.5–25 at.% In; also denoted as ε	Not modelled
Au ₃ In LT	<i>oP8</i> -?-TiCu ₃ <i>Pmmm</i>	$a = 0.58572$ $b = 0.47352$ $c = 0.51504$	Au–In: <339.5	Au–In: 24.5–25 at.% In; also denoted as ε'	(Au) _{0.75} (In) _{0.25}
Au ₉ In ₄	<i>cP52</i> -Al ₄ Cu ₉ <i>P-43 m</i>	$a = 0.984$	Au–In: 482–364.5	Au–In: 28.8–31.4 at.% In; also denoted as γ or Au ₇ In ₃ HT	(Au) _{0.69231} (Au, In) _{0.23077} (In) _{0.07692}
Au ₇ In ₃	<i>hP60</i> -Au ₇ In ₃ <i>P-3</i>	$a = 1.2215$ $c = 0.8509$	Au–In: <374.6	Au–In: 29.8–30.6 at.% In; also denoted as γ'	(Au) _{0.7} (In) _{0.3}
Au ₃ In ₂	<i>hP5</i> -Al ₃ Ni ₂ <i>P-3 m 1</i>	$a = 0.4537$ $c = 0.5659$	Au–In: 457.2–224.3	Au–In: 35.3–39.5 at.% In; also denoted as ψ	(Au) _{0.5} (Au, In) _{0.33333} (In) _{0.16667}
AuIn	<i>aP*</i>	$a = 0.430 \alpha = 90.54$ $b = 1.059 \beta = 90.00$ $c = 0.356 \gamma = 90.17$ $a = 0.65075$	Au–In: <509.6	Au–In: 50–50.1 at.% In Au–In–Sn: solution of about 15 at.% Sn	(Au) _{0.5} (In, Sn) _{0.5}
AuIn ₂	<i>cF12</i> -CaF ₂ <i>Fm-3 m</i>	$a = 0.65124$ $b = 0.65162$ $c = 1.17065$	Au–In: <540.7	Au–In: 66.7 at.% In Au–In–Sn: solution of about 20 at.% Sn	(Au) _{0.33333} (In, Sn) _{0.66667}
Au ₅ Sn	<i>hR6</i> -Au ₅ Sn <i>R3</i>	$a = 0.5092$ $c = 1.4333$	Au–Sn: <190	Au–Sn: 16.7 at.% Sn; also denoted as ζ'	(Au) _{0.84} (Sn) _{0.16}
AuSn	<i>hP4</i> -NiAs <i>P6₃/mmc</i>	$a = 0.43218$ $c = 0.55230$	Au–Sn: <419.3	Au–Sn: 50–50.5 at.% Sn; also denoted as δ Au–In–Sn: solution of about 15 at.% In	(Au) _{0.5} (In, Sn) _{0.5}
AuSn ₂	<i>oP24</i> -AuSn ₂ <i>Pbca</i>	$a = 0.6909$ $b = 0.7037$ $c = 1.1789$	Au–Sn: <309	Au–Sn: 66.7 at.% Sn; also denoted as ε	(Au) _{0.33333} (Sn) _{0.66667}
AuSn ₄	<i>oC20</i> -PtSn ₄ <i>Aba2</i>	$a = 0.65124$ $b = 0.65162$ $c = 1.17065$	Au–Sn: 252–50(?)	Au–Sn: 80 at.% Sn; also denoted as η Au–In–Sn: solution of about 10 at.% In	(Au) _{0.2} (In, Sn) _{0.8}
β -(In, Sn)	<i>tI2</i> -In <i>I4/mmm</i>	In-25 at.% Sn: $a = 0.3459$ $c = 0.4397$	In–Sn: <~150	In–Sn: 12–44 at.% Sn	(In, Sn)
γ -(In, Sn)	<i>hP5</i> <i>P6/mmm</i>	–	In–Sn: <224	In–Sn: 71–97 at.% Sn	(In, Sn)
Au ₄ In ₃ Sn ₃	<i>hP10</i> -Pt ₂ Sn ₃ <i>P6₃/mmc</i>	$a = 0.500$ $c = 1.305$	Au–In–Sn: <~380	Au–In–Sn: at ~40 at.% Au, 30–35 at.% Sn	(Au) _{0.4} (In, Sn) _{0.3} (In, Sn) _{0.3}

2.1. Au–In

The first thermodynamic assessment of the Au–In system was carried out by Ansara and Nabot [2] and subsequently revised by the same authors [3]. More recently, the system has been re-assessed by Liu et al. [4]. In this work, the solid phase equilibria have been improved while keeping, for the liquid, the parameters evaluated by Ansara and Nabot [3], which well reproduce the

temperature dependence of the enthalpy of mixing that has been determined experimentally. Au₄In HT and Au₉In₄ phases have been modelled only by Liu et al. [4], whereas for Au₃In₂, described as stoichiometric by Ansara and Nabot [3], Liu et al. [4] adopted a three-sublattice model, in agreement with the model used by Ansara et al. [5] for the iso-structural phase Al₃Ni₂. Moreover, low temperature phase equilibria in the Au-rich part of the diagram (among the fcc, dhcp and hcp phases) calculated according to Liu et al. [4] seems to be more realistic than presented in the

Download English Version:

<https://daneshyari.com/en/article/1559149>

Download Persian Version:

<https://daneshyari.com/article/1559149>

[Daneshyari.com](https://daneshyari.com)