Chemistry of Metals and Alloys

Chem. Met. Alloys 2 (2009) 30-33
Ivan Franko National University of Lviv
www.chemetal-journal.org

An investigation of the Al-rich region of the Al-Cu-Ir phase diagram

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Received February 2, 2009; accepted June 10, 2009; available on-line November 16, 2009

The Al-Cu-Ir alloy system was studied in the range above 35 at.% Al. Partial 900 and 540°C isothermal sections were determined. Congruent equiatomic AlIr extends at approximately constant Al concentration up to 30 at.% Cu. Cubic C-$Al_{2.7}Ir$ (C-phase) dissolves up to 12 at.% Cu, Al_3Ir up to 8.5 at.% Cu, $Al_{28}Ir_9$ (χ-phase) and $Al_{45}Ir_{13}$ (φ-phase) up to 3 at.% Cu, and Al_9Ir_2 up to 2.5 at.% Cu. Increase of the Cu concentration results in a decrease of the Al concentration of these phases. The Al-Cu ϵ_1-phase dissolves up to 3 at.% Ir, while the solubility of Ir in θ and η_2 is below 0.5 at.%. Close to the high-Cu limit of the C-phase region an fcc C_2-phase (*Fm*$\bar{3}$, *a* = 1.53928 nm) structurally related to the C-phase is formed. A stable decagonal phase (D_1-phase) is formed below 1002ºC in a compositional range extending from $Al_{61.5}Cu_{20}Ir_{18.5}$ to $Al_{59}Cu_{25.5}Ir_{15.5}$. A ternary orthorhombic ε_6-phase (*a* = 2.34 nm, *b* = 1.65 nm, *c* = 1.24 nm) was found below ~1200ºC in a compositional range extending from $Al_{71.5}Cu_6Ir_{22.5}$ to $Al_{68}Cu_{12.5}Ir_{19.5}$. An additional ternary ω-phase (*P*4/*mnc*, *a* = 0.6414 nm, *c* = 1.4842 nm) forming around the $Al_{70}Cu_{20}Ir_{10}$ composition below 683ºC was revealed.

Phase diagram / Intermetallics / Quasicrystals

Introduction

This work continues an investigation of the Al-rich part of the Al-Cu-Ir alloy system, which was carried out in [1], following the first report on the formation of stable quasicrystals in $Al_{65}Cu_{20}Ir_{15}$ [2]. The Al–Cu–Ir alloy system belongs to a group of Al-based systems where quasicrystals and related periodic phases have been extensively studied for two decades (see [3] for references). Also the related ternary alloy systems Al–Cu–Co (see [4,5] and references therein) and Al–Cu–Rh [6] were recently investigated (Co, Rh and Ir belong to the same column of the periodic table). In the present communication we show the results concerning the phase equilibria in a compositional range of above 35 at.% Al at 900°C and in a smaller compositional range close to the Al corner at 540°C. The corresponding partial isothermal sections contain all four ternary phases revealed in [1]. The binary Al-Ir phase diagram was accepted from the recent update in [7], and the binary Al-Cu phase diagram was accepted from [8].

Experimental

Alloys of 3 to 5 g were produced by levitation induction melting in a water-cooled copper crucible under a pure Ar atmosphere. The purity of Al was 99.999%, of Cu 99.95%, and of Ir 99.9%. Parts of solidified ingots were thermally annealed under vacuum at 540°C for 232 h and at 900°C for 186 to 234 h and subsequently water quenched. The as-cast and annealed samples were studied by powder X-ray diffraction (XRD, Cu $K\alpha_1$ radiation was used), scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The local phase compositions were determined in SEM by energy-dispersive X-ray analysis (EDX) on polished unetched cross sections. TEM examinations were carried out on powdered materials dispersed on grids with carbon film. Differential thermal analysis (DTA) was carried out for selected samples at heating and cooling rates of 10 to 50°C/min.

Results

The Al-Ir alloy system [7] contains a number of intermetallics (see Table 1). Congruent AlIr (β-phase) and congruent C-$Al_{2.7}Ir$ (C-phase) melt at temperatures above 2000 and 1600°C, respectively. At higher Al concentration the Al_3Ir, $Al_{28}Ir_9$ (χ-phase), $Al_{45}Ir_{13}$ (φ-phase) and Al_9Ir_2 phases are formed by subsequent peritectic reactions at 1466, 1446, 993 and 877°C. Finally, an (Al) + Al_9Ir_2 eutectic is formed at 657°C.

Table 1 Crystallographic data of the periodic Al-Cu, Al-Ir and Al-Cu-Ir phases mentioned in isothermal sections.

Phase	Space group	Lattice parameters			
		a, nm	*b*, nm	*c*, nm	*β*, °
θ (Al_2Cu)	*I4/mcm*	0.60662	-	0.48738	-
η_2 (AlCu)	*C2/m*	1.2066	0.4105	0.6913	55.04
Al_9Ir_2	$P2_1/c$	0.63779	0.64318	0.87337	94.78
φ ($Al_{45}Ir_{13}$)	*Pnma*	1.6771	1.2327	1.7437	-
χ ($Al_{28}Ir_9$)	*P31c*	1.2286	-	2.7375	-
Al_3Ir	$P6_3/mmc$	0.4246	-	0.7756	-
C ($Al_{2.7}Ir$)	$Pm\bar{3}$	0.7694	-	-	-
β (AlIr)	$Pm\bar{3}m$	0.2969	-	-	-
ε_6 [a]	*Orth.*	2.34	1.65	1.24	-
C_2	$Fm\bar{3}$	1.53928	-	-	-
ω (Al_7Cu_2Ir)	*P4/mnc*	0.64142	-	1.4842	-

[a] Electron diffraction data.

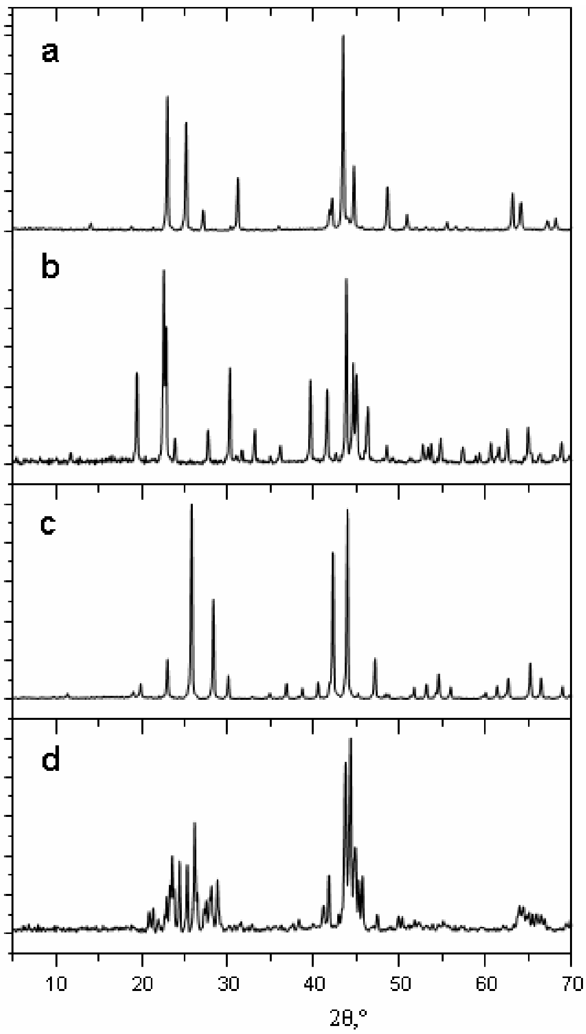

Fig. 1 Powder XRD patterns (Cu $K\alpha_1$ radiation) of the: a) decagonal phase (D_1), b) ω-phase, c) C_2-phase and d) ε_6-phase.

In Al-Cu the phases forming in the relevant compositional range are high-temperature and low-temperatures couples of the ϵ, ζ and η phases and Al_2Cu (θ) [8]. Apart from ϵ_1 they are molten at the temperature of 900°C selected for our major investigation. The θ-phase presented in the partial 540°C isothermal section (see below) is formed by a peritectic reaction at 591ºC. The (Al) + θ eutectic is formed at 548°C [8]. The Al-Cu phases were usually minor in the majority of the studied alloys and were identified according to their compositions compared to those in the binary phase diagram.

In solidified materials equiatomic AlIr was found to extend at approximately constant Al concentration up to 25 at.% Cu. The C-phase dissolves up to 12 at.% Cu, Al_3Ir up to 8.5 at.% Cu, the χ-phase and φ-phase up to 3 at.% Cu, and Al_9Ir_2 up to 2.5 at.% Cu. With the increase of the Cu concentration the compositional ranges of the above-mentioned binary Al-Ir phases, apart from the β-phase, become wider and sharply shifted towards lower Al concentrations. The dissolution of Cu also results in a decrease of the melting temperatures of the Al-Ir phases.

The Al-Cu ϵ_1-phase dissolves up to 3 at.% Ir, while the solubility of Ir in θ and η_2 is below 0.5 at.%.

The formation of a stable ternary decagonal phase and three ternary periodic phases was also observed in the above-mentioned compositional range. Their powder X-ray diffraction patterns are shown in Fig. 1. The decagonal phase of ~$Al_{60}Cu_{24}Ir_{16}$ examined by electron diffraction exhibited a periodicity of ~0.4 nm in the specific direction (D_1-structure, Fig. 2a-c). Of the periodic phases, one was found around the $Al_{70}Cu_{20}Ir_{10}$ composition (ω-phase, see Table 1). It is isostructural to the Al_7Cu_2Rh ω-phase, also observed at similar compositions in Al-Cu-Co(Fe, Ru) [1]. The second ternary C_2-phase (see Table 1) is formed in a range around $Al_{61}Cu_{13}Ir_{26}$, i.e. close to the high-Cu limit of the C-phase compositional range. It exhibits a structure typical of the C_2-phases that are also formed in Al-Cu-Rh and a number of other aluminum-transition metal alloy systems [1]. Around the $Al_{70}Cu_{10}Ir_{20}$ composition, the third ternary phase was revealed, which belongs to the family of so-called ε_i-phases [9]. Electron diffraction examinations in TEM

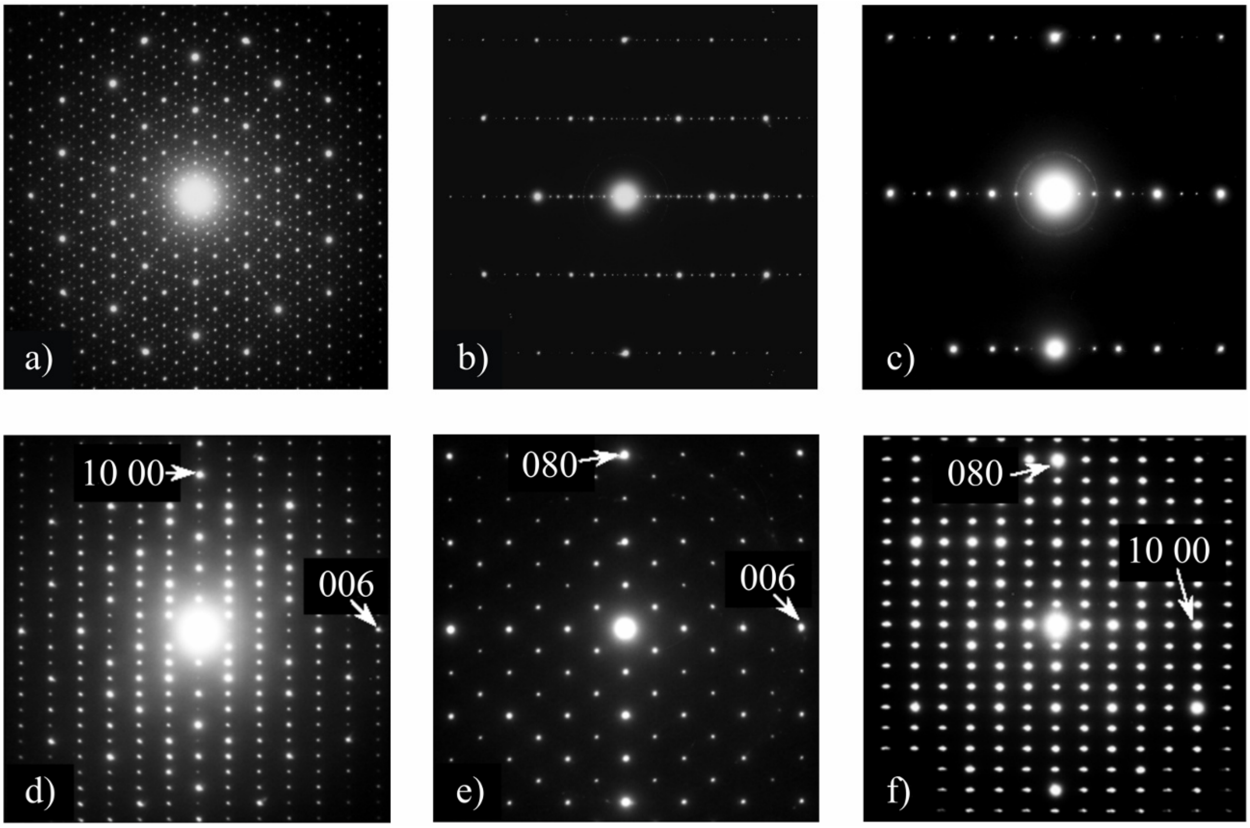

Fig. 2 Electron diffraction patterns of the Al-Cu-Ir phases: a-c) decagonal phase (D_1), d-f) ε_6-phase.

(see Fig. 2d-f) revealed the ε_6 structure, also observed in Al-(Cu)-Rh [6]. While in binary Al-Rh the ε_6-phase is stable and can dissolve up to ~15 at.% Cu, it does not exist as a stable phase in Al-Ir, but is stabilized by Cu in a ternary compositional range.

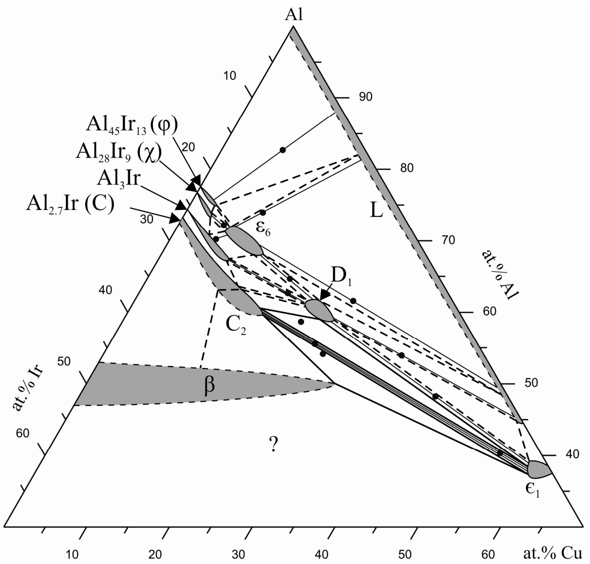

Fig. 3 Partial isothermal section at 900°C. The compositions of the studied samples are shown by spots. Provisional tie-lines are shown by broken lines. L is liquid. The compositional region marked by (?) was not studied.

The D_1-phase melts at about 1002ºC, the ε_6-phase at ~1200ºC, the ω-phase at 683ºC whereas the C_2-phase was not melted up to 1400°C.

The partial 900°C isothermal section of Al-Cu-Ir is shown in Fig. 3. In the studied compositional range the Al-Ir phases β, C, Al_3Ir, χ and φ are solid at this temperature, while a wide region adjacent to Al-Cu is occupied by the liquid and only the ϵ_1-phase is solid. The ternary ε_6-phase is formed in a compositional range from $Al_{71.5}Cu_6Ir_{22.5}$ to $Al_{68}Cu_{12.5}Ir_{19.5}$. The D_1-phase is also solid at this temperature and is formed in a compositional range from $Al_{61.5}Cu_{20}Ir_{18.5}$ to $Al_{59}Cu_{25.5}Ir_{15.5}$. The compositional gap between the existence regions of the C-phase and C_2-phase was not detected and the separation of their compositional ranges is shown conditionally in Fig. 3 by a broken line. It is worth noting that in the Al-Cu-Rh [6] and Al-Pd-Rh [10] alloy systems similar C and C_2 phases were found to coexist in narrow ranges, which was very difficult to detect. The three-phase equilibria Al_3Ir-ε_6-χ, Al_3Ir-ε_6-D_1, L-ε_6-D_1 and L-ϵ_1-D_1 were established with certainty from the existence of the corresponding boundary two-phase equilibria. The three-phase equilibria between the φ, χ, ε_6 phases and the liquid have not been clarified yet due to difficulties to obtain samples in the corresponding small compositional ranges. In Fig. 3 one of the possibilities is assumed.

The partial 540°C isothermal section is shown in Fig. 4. At this temperature equilibration of only a few alloys adjacent to the Al corner was achieved after a

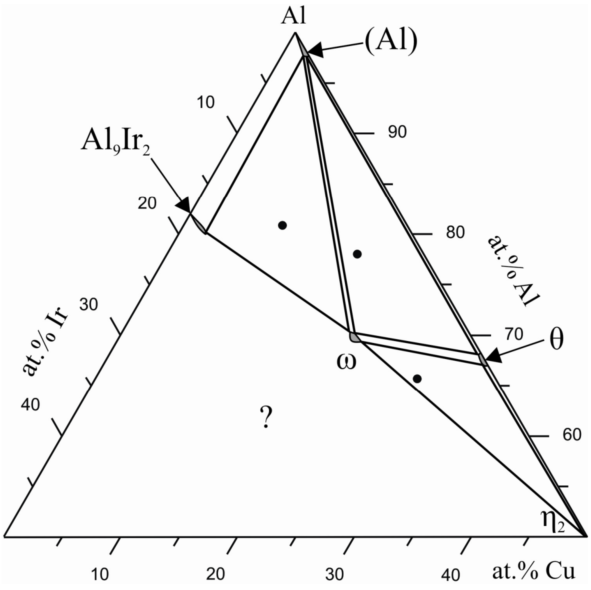

Fig. 4 Partial isothermal section at 540°C. The compositions of the studied samples are shown by spots. Provisional tie-lines are shown by broken lines. L is liquid. The compositional region marked by (?) was not studied.

reasonable time. At this temperature the ternary ω-phase is in equilibrium with the Al_9Ir_2, (Al), θ and η_2 phases.

Conclusions

We report a first study of the Al-Cu-Ir alloy system in the compositional range above 35 at.% Al. Partial 900 and 540°C isothermal sections were determined. The Al-Ir phases were found to dissolve Cu: 30 at.% for AlIr, 12, 8.5, 3, 3 and 2.5 at.% for $Al_{2.7}Ir$, Al_3Ir, χ, φ and Al_9Ir_2, respectively. One stable ternary decagonal phase (D_1-phase) and three ternary periodic phases designated C_2, ε_6 and ω were revealed.

Acknowledgements

We thank V. Lenzen and M. Schmidt for technical contributions. D. Kapush and T.Ya. Velikanova thank Forschungszentrum Jülich for financial support.

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