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Coupled growth of Al-Al_2Cu eutectics in Al-Cu-Ag alloys

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Abstract. Coupled eutectic growth of Al and Al_2Cu was investigated in univariant Al-Cu-Ag alloys during solidification with planar and cellular morphology. Experiments reveal the dynamic selection of small spacings, below the minimum undercooling spacing and show that distinct morphological features pertain to nearly isotropic or anisotropic Al-Al_2Cu interfaces.

1. Introduction

Al-Al_2Cu eutectics in Al-Cu and univariant Al-Cu-Ag alloys are interesting candidates for the investigation of coupled growth patterns because they offer moderate melting temperatures and nearly equal phase fractions in the eutectic solid. More importantly, lamellar coupled growth is regular, since both phases display non-facetted solid-liquid interfaces. Within this paper we present and discuss coupled growth of Al and Al_2Cu in bulk, polycrystalline Al-Cu-Ag samples with planar and cellular solidification morphology. Based on microstructure observations and crystal orientation measurements by EBSD we point out the role of anisotropic interfacial properties that make these alloys even more attractive from today's research perspective.

2. Unidirectional solidification of bulk, polycrystalline samples

Two ternary alloys Al-17.5Cu-1.0Ag, Al-16.0Cu-5.0Ag, all at. %, with nearly univariant composition were prepared for unidirectional solidification experiments in a Bridgman furnace, the sample dimensions being ∅ 8 x 165 mm. Thanks to liquid metal cooling the temperature gradient in the liquid phase close to the solid-liquid interface reached $G=27\pm2\times10^3$ Km^{-1}. Experiments were performed with different, but constant velocity v ranging from 1.0 to 3.1 $\mu m\ s^{-1}$. Within this range the stability limit of planar growth was met, corresponding to constitutional supercooling of the respective alloy at $(G/v)\cong10.4\times10^9$ Ksm^{-2} for 1at.% Ag and $(G/v)\cong27\times10^9$ Ksm^{-2} for 5 at.% Ag. For the given alloys stabilising capillary contributions [11] may be neglected. Quenching, once a length of 40 mm was solidified, finished all experiments. Steady state solidification was safely reached. The as solidified samples were polycrystalline and displayed few eutectic grains at the quenched interface.

Microstructure analysis was carried out at or close to the quenched interface in longitudinal and transverse sections, using an SEM type Gemini 1550 equipped with INCA and INCA Crystal. For EBSD measurements specimens were ion polished as to obtain good Kikuchi patterns for both phases Al and Al_2Cu during crystal orientation mapping. Irrespective of the alloy composition and the solidification velocity, two distinct crystal orientation relationships (ORs) were found between fcc Al and the tetragonal Al_2Cu phase, being characteristic of a given type of grain. Following the systematic classification described in [1], these two ORs and the corresponding grains are called "Beta 6" and "Alpha 4", being summarised in table 1.

The two orientation relationships are adjacent [1] but can be clearly distinguished from one another based on EBSD measurements. It will be shown how the ORs impact on the characteristics of Al-

Al_2Cu eutectic grains, both during planar and cellular solidification, being associated to more or less pronounced anisotropy of the Al-Al_2Cu interfaces. At present, anisotropy effects are being discussed in a qualitative way, since reliable quantitative data on Al-Al_2Cu interfacial properties are lacking.

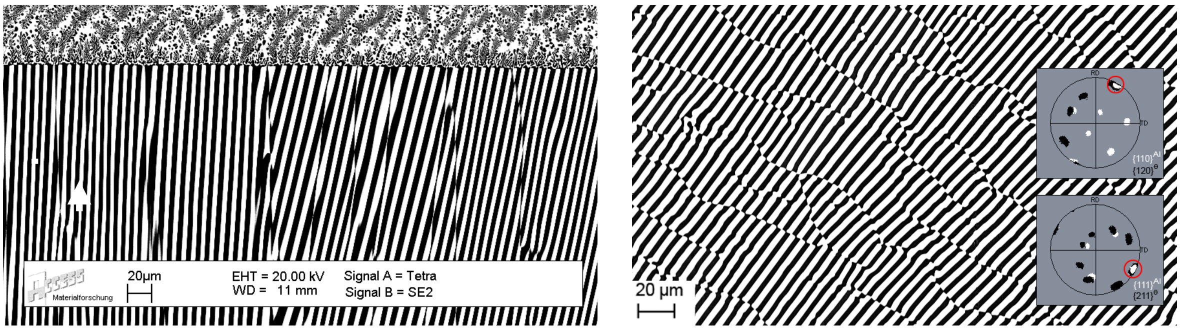
Table 1. Crystal orientation relationships between Al and Al_2Cu in as solidified samples

OR[a]	Common plane	Common direction	Growth direction[b]
Beta 6	$(111)^{Al} // (2\bar{1}1)^{Al2Cu}$	$[\bar{1}10]^{Al} // [120]^{Al2Cu}$	$[9\ 4\ 13]^{Al}, [8\ 7 14]^{Al2Cu}$
Alpha 4	$(130)^{Al} // (100)^{Al2Cu}$	$[001]^{Al} // [001]^{Al2Cu}$	$[001]^{Al}, [001]^{Al2Cu}$

[a] Beta 6 is frequently observed (≈ 75% of eutectic grains), often with tilted lamellae
[b] If the common plane is well aligned to the direction of solidification

3. Lamellar coupled growth and spacing selection in Al-17.5Cu-1.0Ag

Alloy Al-17.5Cu-1.0Ag was selected for the analysis of lamellar coupled growth during solidification with planar solid-liquid interface, aiming to evaluate lamellar spacing selection and to compare it with literature data [2, 3] and model predictions. For the latter, the Jackson-Hunt model [4] and its recent amendment [5] were applied. Lamellar spacing distribution was evaluated in “Beta 6” grains only, more specifically in selected “Beta 6” grains with lamellar interfaces nearly aligned to the direction of solidification. Lamellae tilt angles typically were < 5° and always <10°. By means of example, figure 1 shows one selected grain in longitudinal and transverse section, along with the pole figures of the corresponding OR. The grain and the direction of the transverse cut are marked with a white arrow.



(a) (b)

Figure 1. Eutectic grains with OR “Beta 6” at the quenched interface (a) of a sample from alloy Al-17.5Cu-1.0Ag (at.%) after unidirectional solidification at $v=2.43\ \mu ms^{-1}$ and $G=27\times10^3\ Km^{-1}$. The grain marked with a white arrow (a) is shown in transverse section (b) along with the relevant, superposed pole figures for Al and Al_2Cu.

Lamellar spacings were evaluated for 5 samples solidified at different velocity using image analysis with a resolution of 0,2 μm/pixel. Small measurement areas of about 200x200 μm^2 yielded representative results on spacing distribution, indicating that spacing variations occur within small regions, typically encompassing several fault lines [6]. Table 2 summarises the experimental results. Given are the average spacing λ_{av}, the standard deviation σ_λ, and the minimum and maximum spacing. The table also contains calculated spacing values, λ_m and λ_c, corresponding to the Jackson-Hunt model [4] and its recent amendment [5], respectively. The properties of the different materials used for calculation are listed in table 3. It is assumed that Ag additions of ≤ 1 at.% have negligible impact on solid-liquid interface properties. The lower unvariant eutectic temperature (solidus=531°C) is accounted for with regard to the diffusion coefficient of Cu in the liquid.

Measured spacings compare well with those reported for binary Al-17.4Cu samples that were solidified in capillaries with 0.8 mm diameter [2] at equally low solidification velocities. Indeed, the univariant Al-Al_2Cu eutectic in Al-17.5Cu-1.0Ag behaves much like the invariant binary eutectic regarding spacing selection, at least for this low Ag-content.

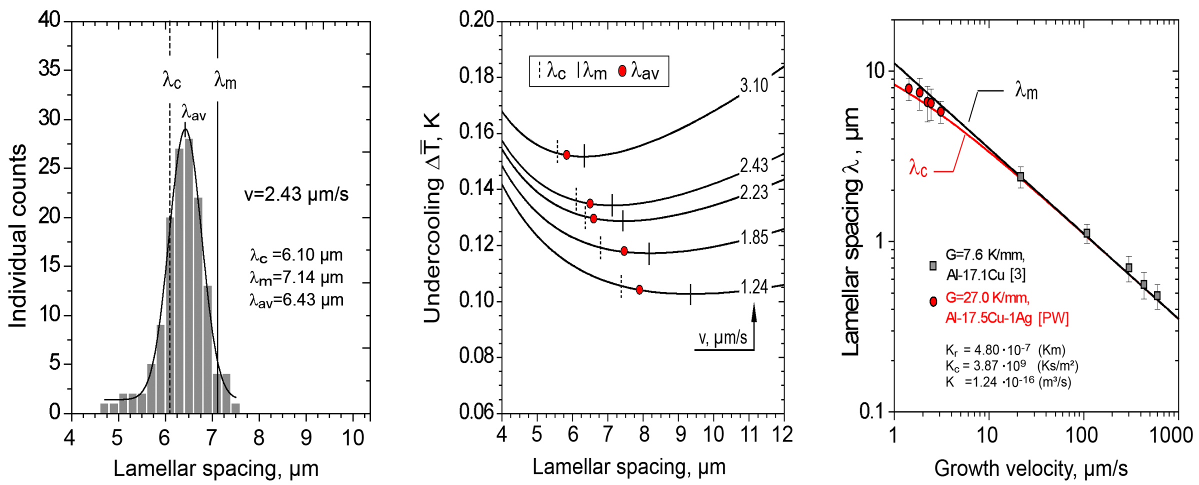
Table 2. Lamellar spacings for alloy Al-17.5Cu-1.0Ag, solidified under G=27x10^3 Km^{-1}

Experiment		Measured spacings			Calculated spacings	
Sample no.	Velocity (μm/s)	$\lambda_{av}\pm\sigma_\lambda$ (μm)	λ_{min} (μm)	λ_{max} (μm)	λ_m (μm)	λ_c (μm)
1	1.42	7.9±0.5	5.8	9.0	9.3	7.4
2	1.85	7.5±0.6	5.6	8.8	8.2	6.8
3	2.23	6.6±0.5	5.2	8.3	7.5	6.3
4	2.43	6.5±0.4	4.8	7.5	7.1	6.1
5[a]	3.10	5.7±0.4	4.8	6.5	6.3	5.6

[a] Sample no. 5 contained 0.8 at.% Ag, allowing to achieve planar growth at the limit of stablity

Fitting the experimentally determined values of λ_{av} according to $\lambda^2 v=K$, the Jackson-Hunt relationship for growth at minimum undercooling [4], leads to a constant K=98.5±3 $\mu m^3 s^{-1}$. However, the calculated value of K, a constant that depends on thermophysical properties only, is K=124 $\mu m^3 s^{-1}$. The difference is significant and indicates that the experimentally selected spacings range below the Jackson-Hunt spacing at minimum undercooling in all the solidified samples.

The recently proposed overstability of small spacings [5] can potentially explain above observations: the smallest stable spacing λ_c, constituting the lower bound of the basic growth mode and corresponding to lamellae pinch-off, is smaller than λ_m, the Jackson-Hunt spacing at minimum undercooling, its value depending on the (G/v) ratio of the given solidification experiment. Both, λ_m and λ_c were calculated following [4, 5] and compared with the experimentally observed lamellar spacings, the key results being illustrated in figure 2. Since it is impossible to achieve planar coupled growth of the univariant alloy Al-17.5Cu-1.0Ag at low (G/v) ratios, literature data [3] reported for binary Al-17.1Cu were chosen instead and included in figure 2(c). Unfortunately ORs were not specified in [3], but are likely to be "Beta 6"as well, this being the most frequently observed OR, at least in our samples.



(a) (b) (c)

Figure 2. Lamellar spacings in alloy Al-17.5Cu-1.0Ag, solidified under G=27x10^3 Km^{-1}: (a) spacing distribution for v=2.42 μms^{-1}, (b) average spacings superposed to Jackson-Hunt growth curves and (c) Al-Al_2Cu eutectic spacings as function of growth velocity for low [3]and high (G/v) ratios.

The results suggest that the λ_c-model [5] can fairly well describe the experimental observations for bulk Al-Al_2Cu eutectics. It is essential to attribute this to continuous processes of lamellae elimination and generation of new lamellae, such that the average spacing is reached and maintained dynamically. Fault lines play a key role for this dynamics [6].

Table 3. Materials properties used for calculation of lamellar spacings in Al-17.5Cu-1.0Ag

Materials property, unit	Al or Al-Liquid	Al_2Cu or Al_2Cu-Liquid	Reference
Phase composition, Cu at.%	2.9	31.9	[6]
Liquidus slope, K/at.%	-7.51	5.45	[6]
Contact angle, °	70	52	[7]
Gibbs-Thomson coefficient, Km	2.4×10^{-7}	5.5×10^{-8}	[7], [8]
Phase fraction in the eutectic	0.5	0.5	[6]
Cu diffusion coefficient (liquid), m^2s^{-1}	3.2×10^{-9} (at T=531°C, extrapolated)		[9]

However, the λ_c-model [5] includes a semi-empirical parameter A, with A=0.15 for 2D axisymmetric eutectics with isotropic interfacial properties, while Al-Al_2Cu interfaces are to some unknown extent, anisotropic. Anisotropy is thought to be strong for the "Beta 6" OR, but weak for the "Alpha 4" OR. This can be inferred not only from lamellar tilt as such, but also from the distribution of misoriented lamellar domains: within a "Beta 6" grain misorientations occur simultaneously in both phases Al and Al_2Cu, leading to a block-like appearance of domains, while this is not the case in "Alpha 4" grains. Figure 3 illustrates this difference; it also shows that fault lines are not always domain boundaries. The presence and local arrangement of misoriented domains depends on the history of grain selection and is not subject of this paper.

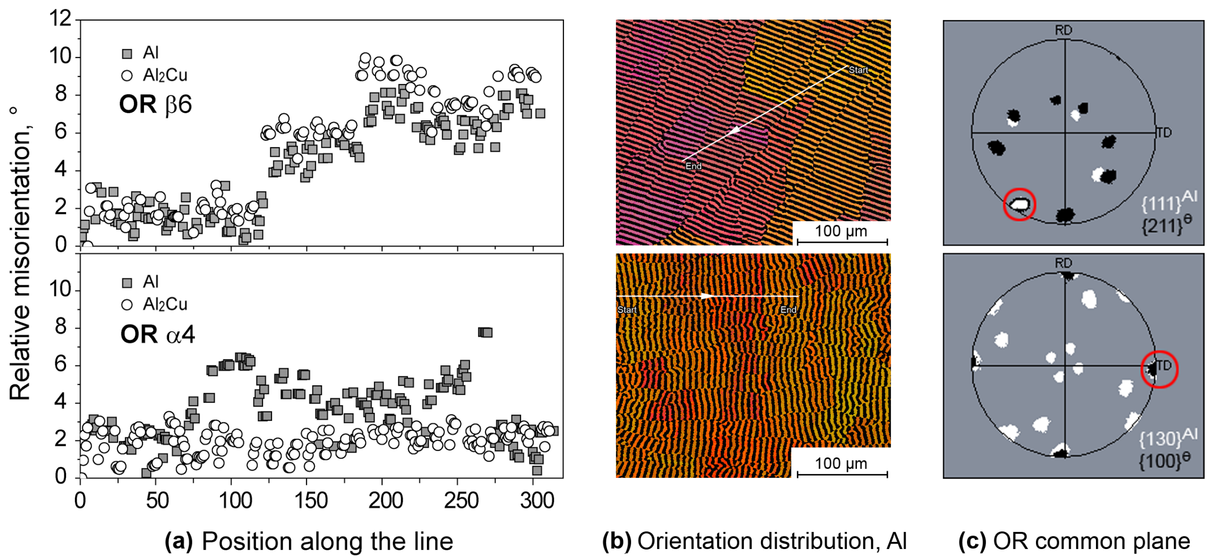
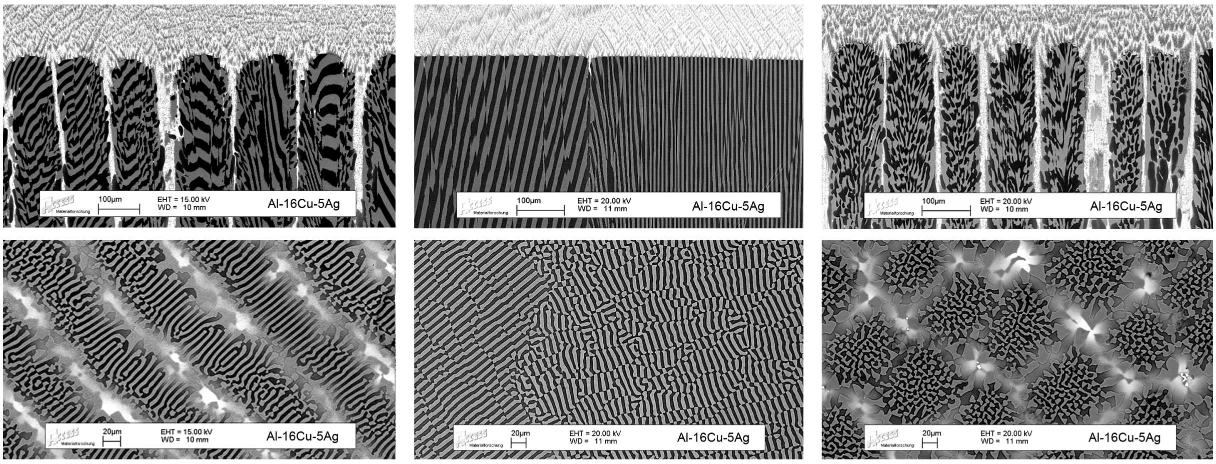


Figure 3. Eutectic grains can accommodate misoriented domains with misorientation angles reaching up to 10°: misorientation angles are shown in (a) along the lines marked in (b) and relative to starting point. Orientation maps in (b) are given for Al in transverse direction, as to guide the eye. The pole figures (c) show the common crystallographic plane that was measured in the EBSD mapping area.

The role of anisotropy is more evidently revealed during coupled growth of Al and Al_2Cu with cellular morphology above the stability limit of planar growth, e.g. above the constitutional supercooling limit. Morphological features of eutectic cells are common to all investigated alloys, while the constitutional supercooling $(G/v)_{crit}$ [10] sensitively depends on alloy composition [6].

4. Cellular coupled growth in Al-16.0Cu-5.0Ag

For alloy Al-16.0Cu-5.0Ag the stability limit of planar growth is lowest compared to all other univariant Al-Al_2Cu eutectics in the ternary Al-Cu-Ag system, due to a particular combination of thermodynamic properties. $(G/v)_{crit}$ was calculated to range around $27{\times}10^9$ Ksm^{-2}, leading to a critical velocity of $v_{crit} \cong 1 \mu ms^{-1}$ for the given temperature gradient. Unidirectional solidification with v=1.6 and 2.2 μm/s leads to deep cells that unlike eutectic colonies [11] are stable morphological patterns at the solid-liquid interface. Two types of cells are found in the samples, being termed elongated and regular cells, as shown in figure 4.



(a) v=1.6 μm/s **(b)** v=1.0 μm/s **(c)** v=2.2 μm/s

Figure 4. Eutectic patterns at the quenched solid-liquid interface of alloy Al-16.0Cu-5.0Ag (at.%) grown at different velocity at $G=27{\times}10^3$ Km^{-1}. Elongated cells (a) and regular cells (c) pertain to grains with OR "Beta 6" and OR "Alpha 4", respectively. Cell boundaries appear bright due to Ag segregation.

The intracellular eutectic retains lamellar character in elongated cells, but appears random in regular cells with preference to align perpendicular to cell boundaries. EBSD measurements yield more insight into the origin of these features, as shown by means of example in figure 5.

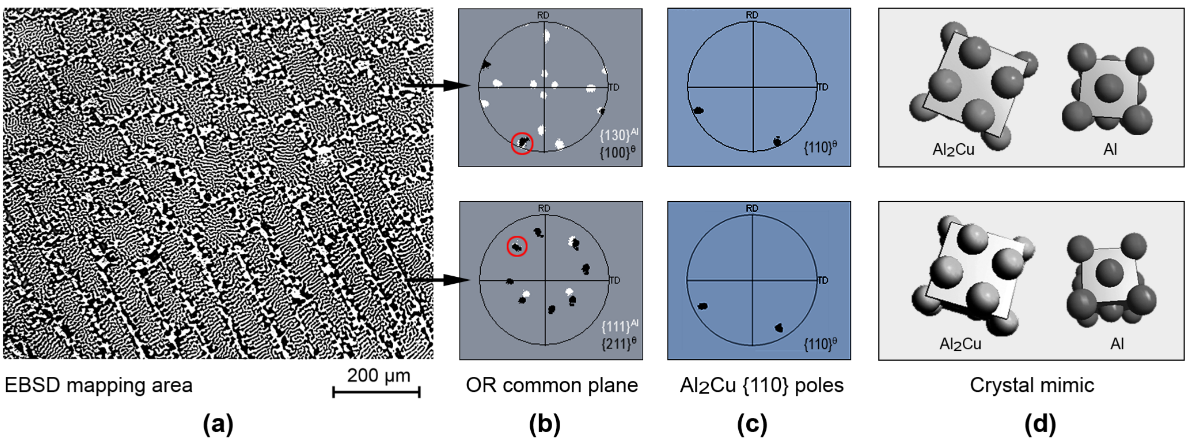


Figure 5. EBSD mapping in transverse section through a sample from alloy Al-16.0Cu-5.0Ag (at.%) solidified at v=2.0 μms^{-1}. Regular and elongated eutectic cells (a) correspond to the "Beta 6" and "Alpha 4" OR (b), respectively. Cell boundaries are aligned to {110}-planes of Al_2Cu (c). For convenience the crystal mimic is shown in (d).

The following correlations between crystal orientation and cellular morphology are remarkable:

(i) in regular cells the "Alpha 4" OR is clearly present, but Al-Al_2Cu interfaces do not follow the common crystallographic plane, being virtually independent from the OR. They tend to align perpendicular to the cell boundaries, both at side faces and at the curved cellular tips (figure 4(c)). This indicates that the interfacial properties of Al-Al_2Cu interfaces in Alpha 4 OR are weakly anisotropic. Under these conditions, cells are free to develop side faces along two {110} planes of the Al_2Cu phase. As described in [12] the {110} planes are associated with lowest attachment energy, being dominant facets during Al_2Cu growth from the melt.

(ii) in elongated cells the "Beta 6" OR is clearly present and the Al-Al_2Cu interfaces remain attached to this OR. This indicates that anisotropy of Al-Al_2Cu interfaces in Beta 6 OR is significant. With this handicap, regular cell formation is impossible and cells remain elongated with boundaries aligned along one {110} plane of the Al_2Cu phase, namely along the one that is nearly but not exactly perpendicular to the lamellae. Whether lamellar tilt relative to the direction of solidification plays an additional role, is yet to be analyzed.

Above observations show that cellular growth is well suited to discriminate between isotropic or anisotropic properties of eutectic hetero-interfaces. However, the quantitative analysis of Al-Al_2Cu interfacial properties remains a topic of future work: grooving techniques [13] combined with EBSD as well as molecular dynamics simulations may allow constructing the Wulff-plot for various ORs.

5. Conclusions and outlook

Univariant Al-Al_2Cu eutectics were grown in Bridgman experiments with either planar or cellular growth morphology. They revealed the overstability of small spacings associated with solidification at high G/v ratios and allowed gaining new insight into the role of Al-Al_2Cu interfacial properties associated with distinct orientation relationships. The experimental observations suggest that the two-phase system Al-Al_2Cu can serve as a good model system for future investigations of anisotropy effects during solidification and perhaps also coarsening. It offers the possibility to select the nearly isotropic or the anisotropic OR "Alpha 4" or "Beta 6", respectively.

Future work will be directed to seeding techniques, aiming to facilitate OR selection. Main emphasis will be placed on repeating and complementing the grooving experiments by Ho and Wheatherly [13], aiming to measure interfacial properties for eutectic Al-Al_2Cu as function of the crystallographic orientation relationship.

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