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COOPERATIVE GROWTH IN THE UNIDIRECTIONALLY SOLIDIFIED
COPPER-CHROMIUM EUTECTIC

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1. Introduction

When a rod-eutectic alloy is unidirectionally solidified the characteristic microstructure is one of rods aligned parallel to the growth direction and arranged on a close packed hexagonal (trigonal) lattice when viewed along the growth direction. Further, if the fibres are faceted with face normals at right angles to the superimposed growth direction, then it is to be expected that all crystallographic variants of the facet lying in the transverse section* are equally likely to occur in a single grain. The electron microscope observations and their analysis reported here, indicate that these expectations are not borne out by the unidirectionally solidified copper-chromium eutectic; the fibres lie with their centres on a distorted hexagonal crystallographic net in the transverse section and within any one grain only one of two crystallographically equivalent facets form on the fibres. These observations suggest that a cooperative phenomenon occurs during the growth of this particular eutectic.

2. Material Preparation.

Master ingots of Cu-Cr were made by melting electrolytic chromium (99.99% purity) and electrolytic copper (99.999% purity) in a magnesia crucible under a residual argon atmosphere using a radio frequency induction furnace.

By using the zone levelling technique⁽¹⁾, the central length was brought to

*In this context a transverse section is one cut at right angles to the ingot growth direction and a longitudinal section is one cut parallel to the growth direction.

the exact eutectic composition. This was found to be close to Cu-1.45wt%Cr; previously reported values⁽²⁾ date from prior to the availability of high purity electrolytic chromium.

Material of eutectic composition was then unidirectionally solidified by either of two techniques. The first was the conventional Bridgman technique with the eutectic contained within a thin quartz crucible and heated by conduction and radiation from a surrounding graphite sleeve in a RF furnace. In the second technique, unidirectional solidification was achieved by Czochralski pulling from a eutectic melt. A growth rate of 110mm/h was used for material prepared by the Czochralski technique, while the growth rate was varied from 600 to 6 mm/h for different specimens grown by the Bridgman method. In all cases the average interfibre spacing, λ , satisfied the diffusion growth equation first proposed for lamella eutectics by Zener⁽³⁾.

$$R\lambda^2 = \text{constant} = 3.5 \times 10^{17} \text{ m}^3/\text{s} \pm 15\%$$

Typically, $\lambda = 1.05\mu\text{m}$ at a growth rate $R = 12.7\text{mm/h}$.

3. Crystallography

After directional solidification the eutectic forms a columnar grain structure with rods of almost pure chromium, 0.1 - 1 μm in cross-sectional width, in a matrix of almost pure polycrystalline copper. When sectioned in the plane normal to the growth direction the fibres are elongated parallelograms with the long facet having a normal $[211]_{\text{Cu}}$, and the shorter sides being rather ill-formed with no distinct facet. Detailed crystallographic studies⁽⁴⁾ indicated that a) the rods are aligned with their long axes within 10^{-1} radians of the imposed ingot growth direction, b) the rod axes are within 10^{-1} radians of $[111]_{\text{Cu}}$ and within $5 \cdot 10^{-2}$ radians of $[0\bar{1}\bar{1}]_{\text{Cu}}$, and

c) that the long facet in the transverse section is within $\pm 5.10^{-2}$ radians of $[211]_{Cu}$.

Furthermore, using a high precision electron microscope micro-diffraction method (4) it was found that in any one grain, each individual fibre satisfied one or other of two crystallographically distinct orientation relationships. One of the orientation relationships was of the Kurdjumov-Sachs type, rotated by $\sim 6 \times 10^{-2}$ radians about the fibre axis, $[111]_{Cr}$, $[0\bar{1}1]_{Cu}$. The other is an "inverted" form of the Nishiyama-Wasserman relation with $(111)_{Cr} || (0\bar{1}1)_{Cu}$; $(\bar{1}01)_{Cr} || (100)_{Cu}$ and $(1\bar{2}1)_{Cr} || (011)_{Cu}$; a relationship obtained from the Kurdjumov-Sachs by a rotation of 9.10^{-2} radians about the common $[0\bar{1}1]_{Cu}$, $[111]_{Cr}$ (fibre axis) direction.

Back reflection Laue x-ray patterns demonstrated that the matrix grains were randomly arranged around the $[0\bar{1}1]_{Cu}$ zone axis.

4. Evidence for Cooperative Growth

The fibre distributions in the transverse sections of the unidirectional solidified eutectic were observed by both transmission electron microscopy and scanning electron microscopy, and the pictures analysed by four separate, but essentially equivalent techniques; direct measurement from the electron micrographs, numerical Fourier Transformation of the fibre coordinates on the micrographs, optical diffractometry using masks prepared from the micrographs, and optical reflection from the selectively etched transverse sections of the eutectic (the optical back-reflection transform method (5)). The latter had the advantage that much larger areas typically 0.1×0.1 mm, could be examined at a time.

These analyses produced the following evidence, indicating that a cooperative mechanism occurred in the growth of the eutectic:

a) In any one grain, scanning electron microscopy shows that every

fibre has its long facet parallel despite the fact that the fibres are aligned close to a $\langle 110 \rangle_{\text{Cu}}$ two-fold axis; $[0\bar{1}1]_{\text{Cu}}$. In other words, only one of the two possible $\langle 211 \rangle_{\text{Cu}}$ facet normals is displayed by the fibres in any one grain.

- b) Although, within any one grain the fibres collectively display both crystallographic orientations relationships with the matrix, only one facet plane is selected, $[211]_{\text{Cu}}$.
- c) The fibres are arranged on a distorted hexagonal (trigonal) net in the transverse section rather than on a close packed hexagonal one. The hexagonal net was distorted such that the angles of the net were, typically, 71, 59, and 50 degrees and the symmetry of the net was reduced to being a diad.

5. Discussion

Although it is not possible to deduce a mechanism from the observation of a microstructure, it is possible to eliminate a number of possible causal mechanisms for the observed cooperative growth, on the basis of symmetry considerations. Since it is observed that the inter-fibre spacing in the transverse section is different in one $\langle 111 \rangle_{\text{Cu}}$ direction than in another, the observed microstructures cannot be the result of either a diffusional or elastic anisotropy, as the diffusivity and stiffness tensors for centrosymmetric crystals such as copper are of fourth rank. In addition, neither a diffusional description nor an elastic anisotropy can explain the presence of a preferred habit variant.

Under appropriate circumstances both a non-symmetric transverse temperature gradient and an external stress could produce an overall alignment of the microstructure. However, it appears unlikely that either is responsible

for the observed features in this particular case. The preferred alignment introduced by a non-symmetric temperature gradient in a polycrystalline alloy would be expected to be continuous across adjacent grains and could be traced without gross discontinuities from one grain to another. Such a behavior was not observed; back reflection optical diffractograms from adjacent grains indicated that the distortion of the fibre lattice was in a different direction in each grain and revealed no tendency for alignment of the distortion direction. In addition, there was no correlation between the long facet direction from one grain to the next. An external stress would similarly create an alignment of the distortion axis and of the long facet direction over all the grains. Neither of these were detected.

Further evidence that precludes either an external stress or an asymmetric transverse temperature gradient as being the causal mechanism is that the back-reflection x-ray diffraction patterns taken of individual grains in the transverse section showed that the grains did not show any systematic arrangement about their $[0\bar{1}1]_{\text{Cu}}$ columnar axes.

On the basis of such considerations it is concluded that during the growth of this eutectic some form of cooperative interaction takes place.

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