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FORMATION OF THE DOUBLETS DURING ORIENTED GROWTH
OF THE Fe-4.34Ni ALLOY

POWSTAWANIE DUBLETÓW
PODCZAS ZORIENTOWANEGO WZROSTU STOPU Fe-4,34Ni

The oriented growth of the Fe-4.34Ni alloy has been performed in the *Bridgman* system. The solid/liquid interface has been frozen and the cellular morphology has been revealed. The doublet cells have been selected from among all the frozen cells. The adequate equation has been applied to describe the geometry of the selected doublet cells. This equation is the solution of differential equation similar to inhomogeneous equation formulated for the pendulum motion with a restraining effect. The shape of revealed doublet cell has been reproduced using the formulated equations. Additionally the doublet shape has been confirmed applying the EBSD technique.

Przeprowadzono krystalizację zorientowaną stopu Fe-4,34Ni w układzie *Bridgmana*. Zamrożono front krystalizacji ujawniając morfologię komórkową. Wśród wielu komórek wyselekcjonowano komórki dubletowe. Dobrano odpowiednie równanie dla opisu ich geometrii. Równanie to jest rozwiązaniem równania różniczkowego analogicznego do niejednorodnego równania opisującego ruch wahadła z hamowaniem. Odtworzono kształt frontu zamrożonej komórki obliczeniami według zaproponowanego równania. Dodatkowo, kształt dubletu potwierdzono na uzyskanej morfologii stosując technikę EBSD.

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1. Introduction

The formation of cellular structure during oriented growth in the 1D experiment or by *Bridgman* system, (2D), is usually treated as a wave character phenomenon, [1]. Moreover, the interplay between different waves appearing at the solid/liquid (s/l) interface should also be taken into account, [2]. The mentioned interplay is based on a periodical function which comprises the fundamental undulation of a formerly planar s/l interface and some disfigurements to the undulation.

The mathematical description of the fundamental undulation of cellular pattern $C_U(x;v)$ comprises not only amplitude $A_U(v)$ and wavelenght $\tilde{\lambda}(v)$ but also primordial shape of the cells. The fundamental undulation of the cellular pattern is given in function of distance x, along which the undulation propagates and growth rate, v, at which crystal grows. The mentioned description was originally made for 1D observation of crystal growing at constant temperature gradient G, measured at the s/l interface. The suggested description is formulated as follows:

$$C_U(x;v) = A_U(v)\frac{[1+Q_U(v)]\sin^2[\pi x/\tilde{\lambda}(v)]}{1+Q_U(v)\sin^2[\pi x/\tilde{\lambda}(v)]}. \tag{1}$$

The equation (1) comprises $Q_U(v)$ – parameter which is responsible for the primordial shape of cells. The 1D observations of cells growth show that four types of cellular pattern can be distinguished: finite cells, doublets, deep cells and gothic cells within predendritic region as revealed by C a r o l i et al. [3] and J a m g o t c h i a n et al. [4].

Calculations of cell shape proved that description of fundamental undulation is not able to reproduce all the details of cell geometry, (for example the s/l interface curvature), [5]. Therefore, equation (1) was modified by the addition of some respective terms of wave character $S_1(x;v)$, which is responsible for certain details of cellular shape curvature. Finally, the total description of cells shape is given as:

$$C_1(x;v) = C_U(x;v) + S_1(x;v). \tag{2}$$

The addition of the $S_1(x;v)$ term does not change the value of the wavelenght $\tilde{\lambda}(v)$ comprised in equation (1). The discussed description is mainly adequate to 1D evolution (*Hele-Shaw's* method) of the cellular pattern but can be adapted in the case of 2D crystal formation (*Bridgman* method) as shown by W o ł c z y ń s k i et al., [6]. The goal of the paper is to reproduce the shape of the frozen s/l interface of doublets revealed due to experiment of directional growth of Fe-4.34Ni alloy.

2. Experiment

The Fe-4.34Ni [at%] alloy has been solidified directionally using the *Bridgman* system. The conditions of experiment were chosen to obtain oriented cellular structure of ferrite. There are: positive temperature gradient about 60 [K/cm], and growth rate 6.6 [μm/s]. A change of the mentioned conditions can lead either to formation of austenite
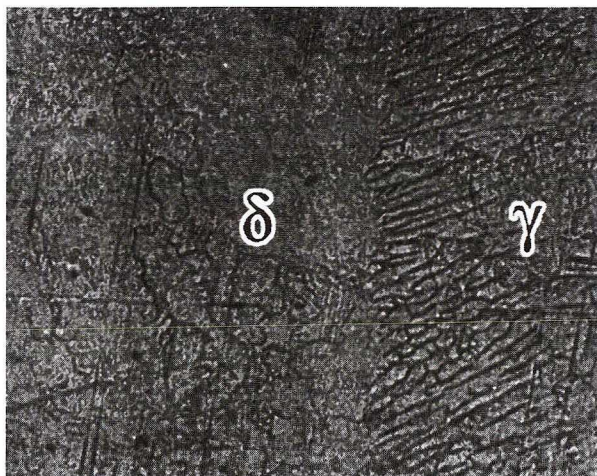
Fig. 1. Formation of ferrite δ or austenite γ for the imposed conditions of solidification; the rapid change of growth rate has been applied at the transition δ/γ, visible within the morphology
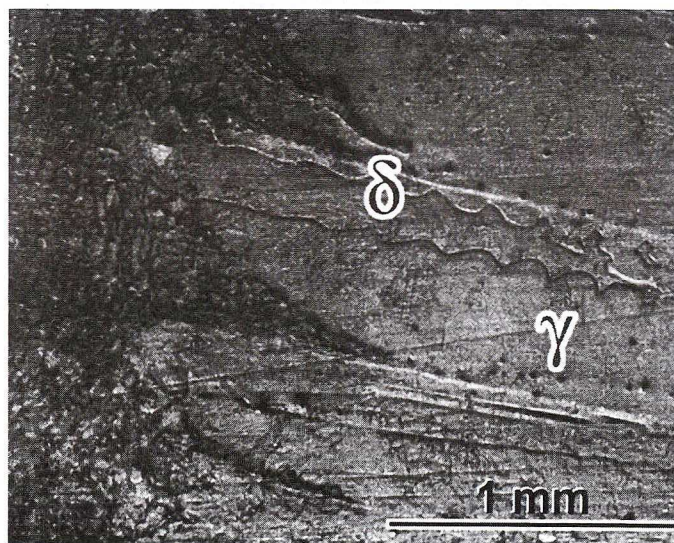

Fig. 2. Competitive growth of ferrite and austenite revealed within growing cell, frozen at a given stage of solidification

or growth ferrite/austenite (δ/γ), Fig. 1. The solid/liquid interface of ferrite/austenite has been frozen and competitive growth between ferrite and austenite has been revealed, Fig. 2, which is described in details by O k a n e et al., [7].

However, in the case when mentioned condition of crystal growth was applied during experiment, the ferrite cells were observed only and instability has been revealed just at its tip. This instability has been described as a first harmonic wave of fundamental
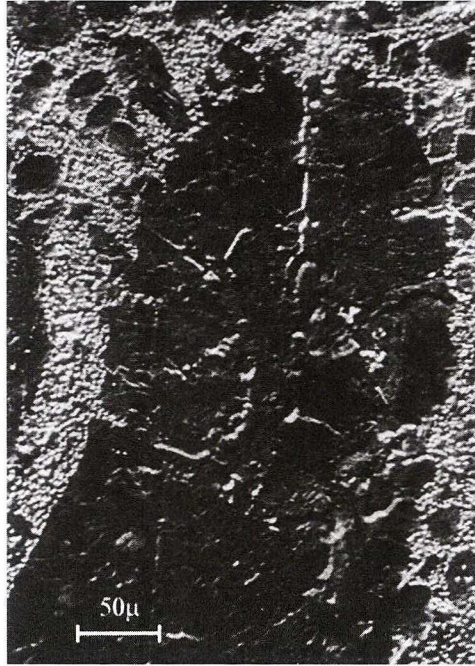
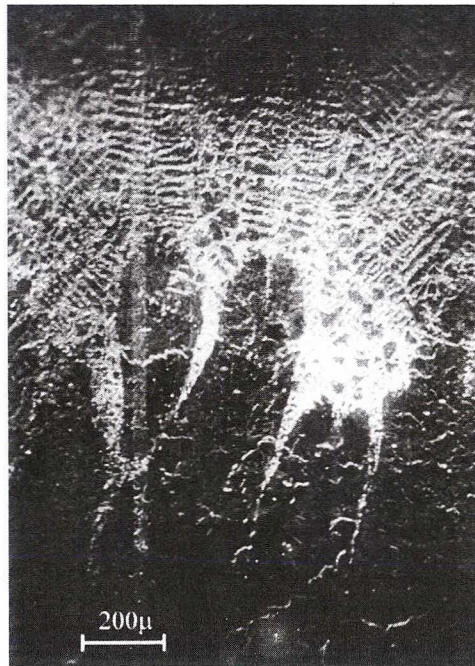Fig. 3. Shape of the doublet solid/liquid interface frozen during directional growth of the δ-ferrite

Fig. 4. Doublets growing within the region of growth rates adequate to deep cells formation of the δ-ferrite, (Fe-4.34Ni)

undulation, which appeared at the formerly planar solid/liquid interface, Fig. 3, Fig. 4. It is to be emphasised that doublets structure is formed for the imposed particular conditions of solidification, only.

3. Description of the doublets formation

The formation of doublets is accompanying by the proper geometry of the solid/liquid interface, Fig. 3. It has been stated that the shape of the s/l interface is to be described using equation (2) in which $S_1(x;v)$ should be formulated in details. The doublets grow within the range of solidification rates adequate to the growth of finite cells or deep cells, shown in Fig. 4. Thus, the equation describing the geometry of s/l interface of doublets are:

a) for the range of growth rates adequate for finite cells formation

$$C_{DF}(x;v) = C_U(x;v) - A_{DF}(v) \frac{[1 + Q_D(v)]\sin^2 \frac{2\pi x}{\bar{\lambda}(v)}}{1 + Q_D(v)\sin^2 \frac{2\pi x}{\bar{\lambda}(v)}}. \tag{3}$$

b) for the range of growth rates adequate for deep cells formation

$$C_{DE}(x;v) = C_D(x;v) - A_{DE}(v) \frac{[1 + Q_D(v)]\sin^2 \frac{2\pi x}{\bar{\lambda}(v)}}{1 + Q_D(v)\sin^2 \frac{2\pi x}{\bar{\lambda}(v)}}. \tag{4}$$
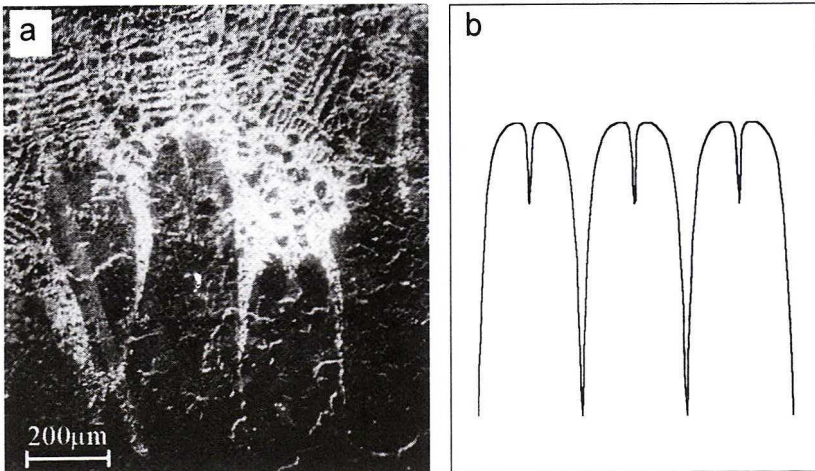

Fig. 5. Doublets of the Fe4.34Ni aloy a) frozen during oriented growth,
b) reproduced due to computer simulation

The equations (4), and (1) were used to reproduce the shape of doublets, as shown in Fig. 5. Equation (1) is responsible for the primordial undulation of previously plane solid/liquid interface.

The second term of the r.h.s. of equation (4) is responsible for asymmetry of the cell tip together with shape of internal groove, that is tip splitting, which corresponds to the harmonic wave.

4. Conlcuding remarks

The geometry of tip splitting of the ferrite doublet has been proved, Fig. 6 applying the EBSD technique, described in details by F a r y n a et al. [8]. However, the Ni-solute redistribution has been measured just below tip splitting of revealed doublet. A result of EDS line-scan measurements in the SEM, Fig. 6 corresponds well to theoretical prediction for redistribution after back-diffusion worked out especially for oriented structure formation (2D crystal growth), [9]. Thus, the measured redistribution can be used to estimate back-diffusion parameter defined by redistribution theory, [9]. Additionally, EBSD technique has been applied to describe the local texture, i.e. a map of single crystallites orientations with respect to the microstructure, Fig. 6.

A unique correlation between fluctuations of the Ni-solute redistribution and crystalline orientations in ferrite δ doublet has been revealed and distinguished by means of vertical line plotted in Fig. 6, (lower part). Moreover, the relation between geometrical asymmetry of doublet and the shape of solute redistribution has also been found, Fig. 6. So, the increase of Ni-solute content visible in the middle of redistribution curve ideally corresponds to the tip splitting of analysed doublet, Fig. 6, (upper part). It should be emphasised that discussed increase of the Ni-solute content in the middle of measurement curve has been revealed in the naighbourhood of the doublet bottom and is not visible far from the doublet bottom. It can be explained by the fact that diffusion within the solid occurs and suppresses the discussed increase.

The crystallites are distributed around the axis of symmetry of explored doublet and each crystallite has own individual angle of inclination to the axis of symmetry. It is because:

1) there are some fluctuations of heat transfer thorough a given doublet; in spite of the fact that generally heat transfer parallel to the axis of symmetry is imposed in the system;

2) some crystallites deflect and turn around their own axis because they have already the crystal structure but properties of the liquid; it occurs in the mushy zone, which exists between liquidus and solidus temperatures.

The vertical dotted lines plotted in Fig. 6 join some boundaries of crystallites with characteristic points of redistribution curve, (with some increases of Ni-solute content). The dashed line joins the increase of Ni-solute content with the splitting of the doublet tip. The full lines join the maximum increase of Ni-solute content with the boundaries of doublet.
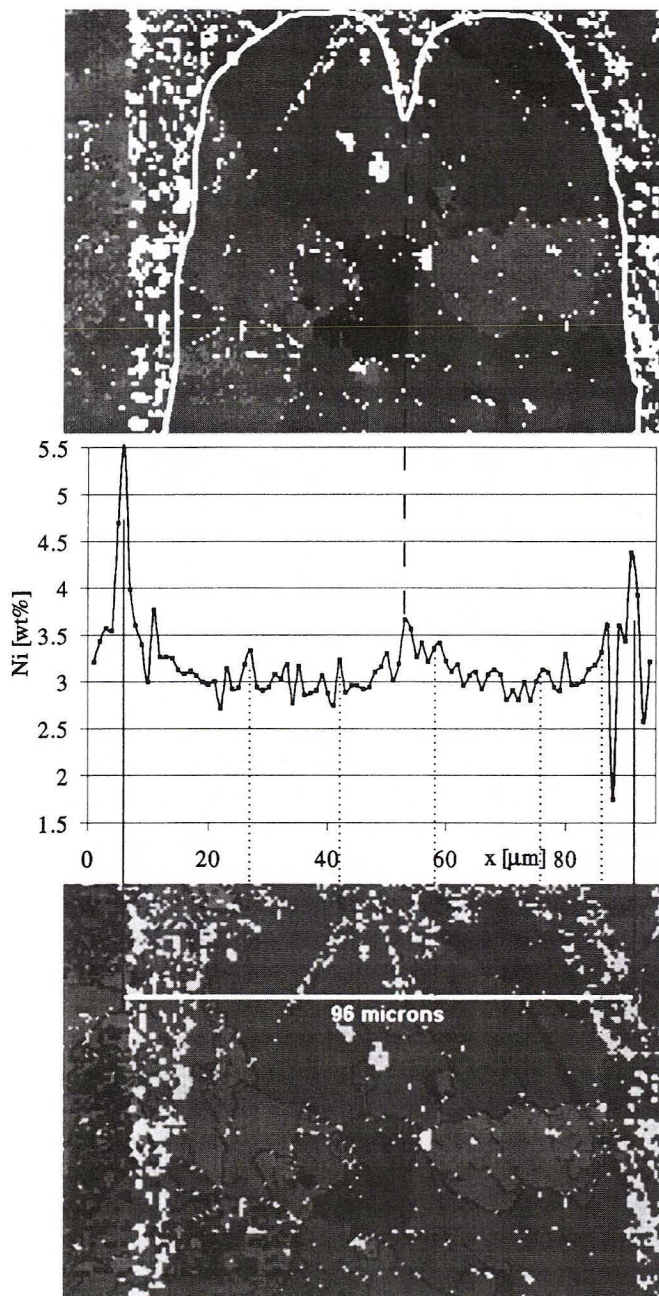
Fig. 6. Map of the sub-crystals orientations within the frozen doublet of the Fe-4.34Ni alloy, (lower picture). The hypothetical shape of the doublets, frozen s/l interface, (upper picture). Some small areas of non-identified orientation are decorating the doublet outside its interface. Ni-solute redistribution related to the asymmetrical shape of doublet, (upper picture) and to sub-crystals boundaries, (lower picture)

Tip splitting as well as doublet asymmetry have been described and reproduced by using equations (4) and (1). The description is suggested as a sum of two similar products. The first component of the sum is responsible for the primordial undulation of the s/l interface, the second for the asymmetry of doublets.

However, equation (1) can be treated as a quotient:

$$C_U(x;v) = \frac{C_{UB}(x,v)}{C_{UC}(x;v)}, \tag{5}$$

where

$$C_{UB}(x;v) = A_U(v) \sin^2 \frac{\pi x}{\bar{\lambda}(v)} \tag{6}$$

$$C_{UC}(x;v) = \frac{1 + Q_U(v) \sin^2 \dfrac{\pi x}{\bar{\lambda}(v)}}{1 + Q_U(v)}. \tag{7}$$

Equation (6) describes the doublets shape resulting from the bifurcation phenomenon of the $A_U(v)$ amplitude. Equation (6) fulfils the following differential equation:

$$\frac{d^2 C_{UB}(x;v)}{dx^2} - 2 \frac{\pi^2}{\bar{\lambda}^2(v)} [A_U(v) - 2 C_{UB}(x;v)] = 0 \tag{8}$$

which is similar to the inhomogeneous differential equation describing the motion of a pendulum with a restraining effect. Analogously, equation (7), which describes the tip splitting, fulfils the following differential equation,

$$\frac{d^2 C_{UC}(x;v)}{dx^2} - 2 \frac{\pi^2}{\bar{\lambda}^2(v)} \left[\frac{Q_U(v) + 2}{1 + Q_U(v)} - 2 C_{UC}(x;v) \right] = 0 \tag{9}$$

which is also similar to the inhomogeneous differential equation describing the motion of a pendulum with a restraining effect

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