



Full length article

# In-situ observation of coupled peritectic growth in a binary organic model alloy



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## ABSTRACT

Peritectic alloys form a variety of solidification morphologies of which peritectic coupled growth (PCG) is the most peculiar. Because the conditions that lead to PCG are still under discussion, in-situ investigations of the evolution from lateral bands to PCG were carried out by using binary organic TRIS-NPG model alloys at concentrations in the peritectic region. The experiments were done vertically with relatively large rectangle glass tubings. For a hyper-peritectic alloy at growth rates below the morphological stability limits of both solid phases, we found that first the properitectic phase grew while lateral bands of the peritectic phase formed between the glass walls and the properitectic phase. Then, the peritectic phase dominated the growth until lateral bands of the now properitectic phase occurred, again close to the glass walls. Now both phases grew side-by-side as two layers at different depths in the sample. However, the properitectic phase spread locally onto the peritectic/liquid interface, so that patches of peritectic phase surrounded by a properitectic matrix developed. This resulted in creating an unstable PCG mode with cycles of increasing and decreasing phase amounts, which from a side-on view resemble tulip- and onion-like structures respectively.

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## 1. Introduction

Many alloys of great industrial significance show a phase diagram with a peritectic reaction (steel, Al- and Cu-alloys, high  $T_c$  superconductors and rare earth permanent magnets) [1]. According to such phase diagrams, peritectic alloys form a primary  $\alpha$ -phase first upon cooling from the melt and then at the peritectic temperature,  $T_p$ , a second solid  $\beta$ -phase called peritectic phase. Above  $T_p$ , the  $\alpha$ -liquid equilibrium is stable and the  $\beta$ -liquid equilibrium is metastable, whereas the opposite is true below  $T_p$ . At  $T_p$ , a melt with concentration,  $c_p$ , is in equilibrium with the primary  $\alpha$ -phase at  $c_{p\alpha}$  and the peritectic  $\beta$ -phase at the so-called peritectic concentration  $c_{p\beta}$ . Alloys with  $c_{p\alpha} < c_0 < c_{p\beta}$  are called hypo-peritectic and those with  $c_{p\beta} < c_0 < c_p$  are called hyper-peritectic. Here,  $c_0$  is the initial alloy concentration.

For conditions where both phases are supposed to grow with a morphologically stable planar solid/liquid interface ( $s/l$  interface), the transient growth process necessary to reach a steady-state growth with a planar  $s/l$  interface of one phase is hindered by

the nucleation and growth of the other phase. This in turn then passes through a transient growth process, hindered by the nucleation and growth of the initial phase. This interplay between solute transport and nucleation kinetics leads to the formation of a banded structure consisting of layers of the two solid phases extended perpendicular to the growth direction [2–6].

A conceptual description of cyclic nucleation and growth under purely diffusive condition was suggested by Trivedi in Ref. [7]. However, Trivedi's ideal concept fails in many real solidification cases for three reasons. First, often the presence of melt convection changes the melt concentration locally as well as at the sample scale [8,9]. Second, the lateral spread of the phase which has just nucleated, competes with the forward growth of the present phase and thus incomplete bands - so-called islands or island banding - form [10–12]. Finally, in real systems, the three dimensionality of an unsteady  $s/l$  interface motion may lead to a dynamic phase interaction where nucleation is of less importance [9]. In bulk samples the dominant growth of one phase can be accompanied by a simultaneous growth of a small amounts of the second phase somewhere in the sample. A sudden intensified appearance of the second phase might then look like nucleation but is in fact a rapid spread over the first phase.

In addition, lamellar- or rod-like peritectic coupled growth

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(PCG), similar to those typically observed in eutectics were found and presented in Refs. [9,13–16]. PCG reportedly only occurs when both primary and peritectic solid phases are morphological stable (cf. reference). Obviously, both PCG and island banding morphologies can form under apparently similar conditions. To investigate the underlying microstructure selection, Lo et al. [9] conducted both corresponding experiments and phase field simulations. They found that island banding may be either sustained in time, or in a transient microstructure that ‘seeds’ coupled growth. If that is the case, two basic conditions must be satisfied: (i) there must exist a stable range of coupled growth spacing; and (ii) the mean distance between islands must fall approximately within this stable range.

In the present paper, we report in-situ observations of the transition from banding to PCG using thick samples (3D) of an organic model system, which reveals a non-faceted/non-faceted two-phase peritectic alloy. The paper is structured by describing the experimental procedure in Section 2 and the corresponding in-situ observations of banding and PCG in Section 3. For a correct identification of the different solid phases growing in the thick samples, we discuss in Section 4 features of the three-dimensionality of the observation, the formation of lateral bands, the transition between cycles of PCG, and finally, how isothermal PCG develops from banding. Conclusions and a summary are given in Section 5.

## 2. Experimental

The solidification experiments were carried out with alloys consisting of two organic components, namely TRIS<sup>1</sup> (Tris(hydroxymethyl)-aminomethane) and NPG (Neopentylglycol).<sup>2</sup> Both components show the presence of transparent high temperature non-faceted phases [17] (also called plastic phases), which are supposed to solidify with similar solidification morphologies as metals. The phase diagram shows a peritectic region from  $c_{pz} = 0.47$  to  $c_p = 0.54$  mol fraction NPG with the peritectic concentration at  $c_{p\beta} = 0.51$  mol fraction. Detailed information on the TRIS-NPG phase diagram as well as on important materials properties of NPG and TRIS can be found in Ref. [18]. Directional solidification experiments were carried out with alloy compositions in steps of 0.1 mol fraction NPG over the whole peritectic range. In this paper we present detailed studies with alloys that had of  $c_0 = 0.52$  mol fraction NPG.

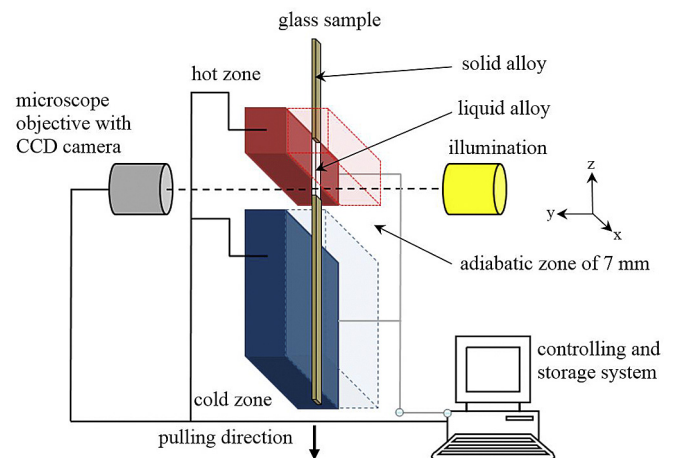
For the investigations, 200 mm long rectangular tubes with a width of 6.8 mm and a depth of 1.4 mm were used. The wall thickness was 0.4 mm so that the inner width and depth of tubes were 6.0 mm and 0.6 mm, respectively. The organic alloys inside the tubes were solidified vertically upwards with a constant pulling rate,  $V_{pull}$ , in a given temperature gradient,  $G$ . The reason for choosing these relatively thick tubes was the fact that post-mortem studies of cylindrical metallic alloy samples showed some three-dimensional features of the formation mechanisms that results in PCG [9,14,16]. These experiments were also performed with a vertical upwards growth direction. The tubes were filled by capillarity under Argon atmosphere of about 80% (around 20 mm at the upper end was left empty) and then hermetically sealed with a glue [18]. Such samples were then placed into the preheated micro Bridgman-furnace and held at least 120 min in rest to establish a thermally equilibrated state and a planar  $s/l$  interface. As a sample is longer than the heated zone, only a sample segment of around 10 mm in length was molten. The pressure increase, which may

originate from the expansion on heating and melting, was released by a solid material column at room temperature not filling the tube completely. The low-temperature faceted phases, which are stable at room temperature, reveal 10–20% higher densities, compared with the high temperature plastic phases; visible gaps and cracks occur as the faceted phase contracts on cooling. Hence, the heated and finally molten segment can easily push the solid segments aside, both at the top and bottom, and a pressure-free molten segment can form. Note that the results presented in this paper were gained by using newly filled samples in which the concentration can be assumed to be uniform along the sample's length. After the preheating period of 120 min, the sample was moved at constant velocity into the cold part of the Bridgman furnace and a digital camera recorded and stored pictures of the  $s/l$  interface with a time interval of  $\Delta t = 30$  s.

The micro Bridgman-furnace was made up of two brass parts fixed on symmetrical ceramic shelters to create a 7 mm adiabatic gap (Fig. 1). The temperature within the brass parts were controlled by electrical resistant heaters and measured with Pt 100 temperature sensors placed inside each brass block. A slot ( $6.5 \times 1.2$  mm<sup>2</sup>) was milled into the brass blocks in which the glass sample could slide. The sample was illuminated through glass windows in the ceramic plates at the adiabatic zone to observe the morphology of the  $s/l$  interface with a ZEISS microscope, in combination with a black-and-white camera. During solidification, images were recorded simultaneously with the relevant temperatures of both brass parts. The movement of the glass sample through the furnace was PC-controlled. Details on alloy preparation, filling and sealing of the sample, as well as on the Bridgman-furnace can be found in Refs. [18–23].

In order to describe the experimental findings in the next section a coordinate system is defined in Fig. 1. The longitudinal direction is taken as z-direction, the wide side of the rectangular tube (width) is taken as x-direction and the small side (depth) as y-direction.

Both non-faceted high temperature solid phases of the TRIS-NPG system, namely the TRIS-rich properitectic  $\alpha$ -phase and the peritectic  $\beta$ -phase, are equally transparent and thus optically indistinguishable. The identification of the solid phases is therefore only possible by careful observation of their growth dynamics. This will be done in the discussion section. In the result section 3, we only name the two solid phases as first and second leaving the exact



**Fig. 1.** Sketch of the micro Bridgman-furnace. The brass parts of the backward ceramic shelter (not shown in the sketch) and the solid alloy within the glass sample are full colored. The brass parts on the front ceramic shelter and the liquid alloy are shown transparent.

<sup>1</sup> <http://en.wikipedia.org/wiki/Tris>.

<sup>2</sup> <http://www.sigmaldrich.com> or <http://www.merck.de>.

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