

## Effects of the initial melt undercooling on solid-state recrystallization and grain growth processes in as-solidified superalloy

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Recently, detailed studies in solidification process [1], microstructure evolution [2], and grain refinement mechanism [3] were carried out for undercooled DD3 (Ni alloyed with 9.5 Cr, 5.9 Al, 2.2 Ti, 5.2 W, 5 Co, and 3.8 Mo, wt%) superalloy melts. If the initial melt undercooling ( $\Delta T$ ) before solidification is above a critical value, 180 K, then the phase transformation consists of three domains [3]: (1) the first mushy zone (FMZ), i.e., rapid solidification of Ni-based  $\gamma$  solid solution during recalescence; (2) the second mushy zone (SMZ), i.e., normal solidification controlled by natural cooling after recalescence; (3) solid-state recrystallization and grain growth processes, as well as the subsequent  $\gamma'$  ( $\text{Ni}_3\text{Al}(\text{Ti})$ ) precipitation [4].

For  $180 \text{ K} < \Delta T < 300 \text{ K}$ , the as-solidified morphology is composed of the refined granular crystals with an average diameter as 20–50  $\mu\text{m}$  [1–3]. We suggested that this kind of grain refinement should be caused by solid-state recrystallization as argued by Powell [5], which appears to occur during, or immediately after, solidification, while the solid metal is very close to the melting point. As would be expected,  $\Delta T$  not only controls the solidification, but it also influences the subsequent solid-state recrystallization and grain growth. In the present letter, we concentrated on recrystallization and grain growth processes associated with high-undercooling rapid solidification, together with an attempt to highlight the effect of  $\Delta T$  on the recrystallized grain size.

In cases where dendrites develop and grow, a coherent dendrite network is usually established at solid fractions between 10 and 30 percent [6]. The dendrite coherency marks a point in FMZ [3] where the material starts to develop strength, and where the resistance to material movement increases drastically. As solid fraction after recalescence further increases with  $\Delta T$  [7], the inter-dendritic permeability is reduced, thus resulting in an increasing pressure gradient within FMZ [8]. The pressure gradient and flow of the liquid means that stresses are created and exchanged between the solid dendrites as well. Therefore, the solidification shrinkage, the thermal contraction of the solid, the dendrite-dendrite interactions in the dendrite network, and the governing pressure differential for flow [8] cause the development of large stresses,  $\sigma$ , in the solid as the flow resistance or solid fraction increases in FMZ [3]. As

illustrated in Fig. 1, the calculated stress develops in the dendrite network as  $\Delta T$  increases [3]. If  $\Delta T > 180 \text{ K}$ , the stresses exerted upon the solid network become higher than its strength, and deformation can probably intervene [9]. This interpretation is compatible with the microstructure evolution as observed in [1–3], where the dendrite network collapses as a result of the large stress arising from rapid solidification.

The rapidly solidified DD3 superalloy could be assumed as a heavily cold-worked material [11], which could transform to its defect-free state by nucleation of new lattice and growth of the nuclei at the expense of the defect-containing parent material. This heterogeneous transformation, in which new crystals are created, is known as recrystallization. Approximately, the Gibbs free energy change,  $\Delta G$ , available for the transformation, equals the enthalpy increase (stored energy,  $E_s$ ) of materials due to cold work, which has been experimentally determined for a number of metals as function of the amount of cold work, deformation process, and strain rate [10]. In the present case, however, it is due to the high strain energy arising from the rapid solidification, but not any other external agency, that results in dendrite distortion, fragmentation, and recrystallization as well. Recognizing that the recrystallization proceeds within a limited temperature range [3, 4, 11], it could be assumed as an isothermal process. Since 60 percent solid fraction remains after recalescence if  $\Delta T > 180 \text{ K}$ , it is reasonable to assume the maximal recalescence temperature  $T_R$  to be the recrystallization temperature [3, 11].

Recrystallization is a nucleation-growth process. For recrystallization occurring in the as-solidified DD3 superalloy, detailed description for nucleation rate,  $\dot{N}$ , and growth rate,  $\dot{G}$ , is available in [11]. Different  $\Delta T_s$  give different  $\dot{N}$  and  $\dot{G}$ , and in turn, different response of the material to recrystallization, i.e., the final grain size is strongly dependent on the response of the material to recrystallization. According to [11, 12], the grain size after isothermal recrystallization and grain growth is proportional to the value of growth-nucleation ratio  $(\dot{G}/\dot{N})^{1/4}$  (here defined as  $\dot{G} - \dot{N}$  ratio). Substituting the analytical descriptions for  $\dot{N}$  and  $\dot{G}$  into  $\dot{G} - \dot{N}$  ratio gives quantitative analysis between  $\dot{G} - \dot{N}$  ratio and  $\Delta T$ , as well as that between the final grain size and  $\dot{G} - \dot{N}$  ratio, as shown in Fig. 2a and b, respectively.

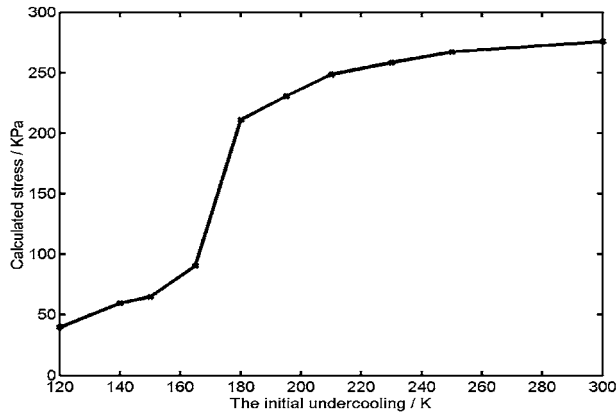


Figure 1 Stress development in rapid solidification of DD3 superalloy as a function of the initial melt undercooling.

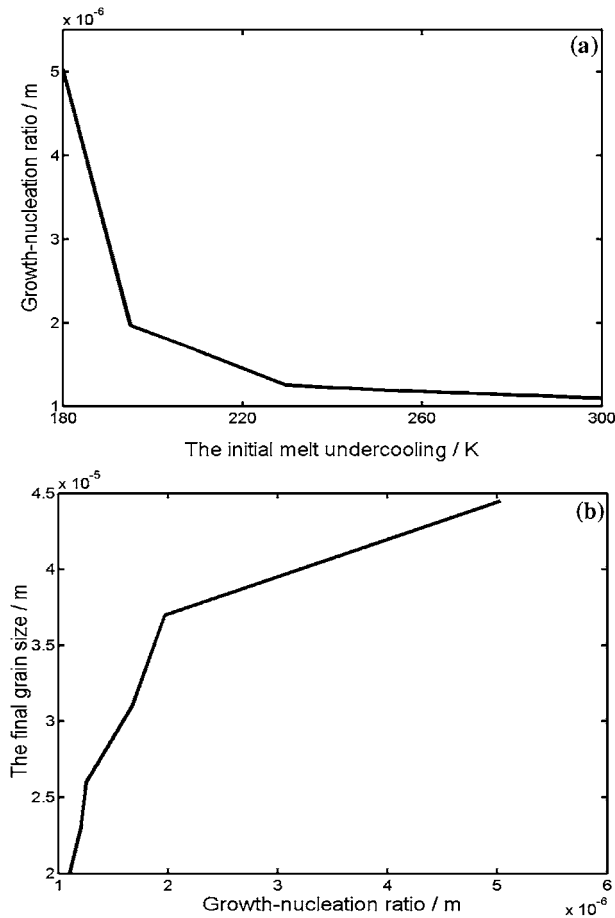


Figure 2 Relation between: (a) the initial melt undercooling and  $\dot{G} - \dot{N}$  ratio and (b)  $\dot{G} - \dot{N}$  ratio and the final grain size, obtained in DD3 superalloy subject to high-undercooling rapid solidification and solid-state transformation.

With increasing  $\Delta T$ , the stored energy is expanded due to stress development (see Fig. 1), and the change in the nucleation rate becomes much more than that in growth rate [11], thus the value of  $\dot{G} - \dot{N}$  ratio decreases (see Fig. 2a). Accordingly, smaller grain sizes are obtained (see Fig. 2b). This indicates that the final grain size after recrystallization and grain growth occurring in the as-solidified matrix varies inversely with  $\Delta T$ , as confirmed by the corresponding microstructure evolution [3]. Nevertheless, the slope of the curve increases with the reduction of  $\dot{G} - \dot{N}$  ratio, i.e., the increase of  $\Delta T$ , a

clear proportional relationship between grain size and  $\dot{G} - \dot{N}$  ratio [11, 12] does not hold over the whole undercooling range (see Fig. 2b).

According to [11], the final grain size subjected to the isothermal grain growth is determined by activation energy for grain growth,  $Q$ , the grain growth time,  $t$ , and the grain boundary (GB) energy,  $\sigma_b$ . In the present case, grain growth proceeds by natural cooling after recalescence [3–5], so it is reasonable to assume  $Q$  and  $t$  as constants. Hence the only factor that determines the final grain size should be  $\sigma_b$  [11]. According to the Gibbs adsorption equation [13] and the thermodynamic approach of Weissmüller [14] and Kirchheim [15],  $\sigma_b$  is reduced by solute segregation, and if it is possible to reduce  $\sigma_b$  to zero, then there would be no driving force for grain growth in such a binary polycrystalline closed system. Under the critical assumptions: (1) segregation is of the saturation type, with a finite number density of segregation sites; (2) the number of solute and solvent atoms is conserved, and (3) precipitation of an intermetallic compound must be suppressed, the following equation is deduced [14, 15],

$$\sigma_b = \sigma_0 - G(\Delta H_{\text{seg}} + RT \ln(X_0)) \quad (1)$$

with  $\sigma_0$  as the GB energy for pure solvent,  $G$  as the solute excess at the GBs at saturation,  $\Delta H_{\text{seg}}$  as the segregation enthalpy change per mole solute (here defined as being positive for GB enrichment),  $T$  as  $T_R$ , and  $X_0$  as the solute content within the grains. The second term in brackets in Equation 1 describes the loss of configurational entropy caused by segregation. Since  $\Delta H_{\text{seg}}$  is gained by enriching solute atoms at the GBs,  $\sigma_b$  should decrease from its original value  $\sigma_0$ .

In the normal solidification after recalescence, the inevitable equilibrium segregation makes the alloying elements of DD3 superalloy be concentrated on the GBs in the as-solidified  $\gamma$  solid solution [1–3]. Suppose that  $\gamma'$  precipitation does not coincide with grain growth,  $\Upsilon$  solid solution could be considered as an ideal binary polycrystalline closed system with constant  $\Delta H_{\text{seg}}$ . Because of the large grain size (i.e., in the  $\mu\text{m}$  range) [3], it is reasonable to assume that  $G$  has reached its saturation value, independent of  $\Delta T$ . With increasing  $\Delta T$ ,  $T_R$  is reduced, in contrast with an increased  $X_0$  due to solute trapping effect [1–3], so a greater reduction happens to the GB energy according to Equation 1. Higher  $\Delta T$  leads to lower GB energy, and subsequently, insufficient grain growth or further grain size decrease takes place. This explains why the same change in  $\dot{G} - \dot{N}$  ratio gives more decrease in grain size, as  $\Delta T$  increases (see Fig. 2b).

In summary,  $\Delta T$  not only determines the solidification process, but its heredity will also be remained to influence the subsequent solid-state recrystallization and grain growth processes. The final grain size is strongly dependent on the response of material to recrystallization. With increasing  $\Delta T$ , rapid solidification produces larger strain energy, provides stronger response to recrystallization, and thus leads to smaller grain size of the as-solidified materials. In connection with the thermodynamical approach developed by Weissmüller

and Kirchheim, the much smaller grain size associated with higher  $\Delta T$  can be ascribed to insufficient grain growth in the recrystallized matrix owing to a significant reduction of GB energy.

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### References

1. F. LIU and G. C. YANG, *Metal. Mater. Trans. B* **32** (2001) 449.
2. *Idem.*, *Mater. Sci. Eng. A* **291** (2000) 9.
3. *Idem.*, *J. Cryst. Growth* **231** (2001) 295.
4. *Idem.*, *Mater. Trans.* **42**(6) (2001) 1135.
5. G. L. F. POWELL and L. M. HOGAN, *Trans. TMS-AIME* **242** (1968) 2133.

6. C. J. PARADIS, L. AMBERG and H. J. THEVIK, A. MO, 1995, *Modeling of Casting*.
7. T. J. PICCONE, Y. WU, Y. SHIOHARA and M. C. FLEMINGS, *Mater. Trans. A* **18** (1987) 925.
8. "Welding and Advanced Solidification Processes VII," edited by M. Cross and J. Campbell (TMS, Warrendale, PA) p. 609.
9. J. PILLINGS and A. HELLA WEL, *Metall. Trans. A* **27** (1996) 229.
10. K. AUST and J. RUTTER, "Recovery and Recrystallization of Metals," edited by L. Himmel (Gordon and Breach Science Publishers, New York, 1963).
11. F. LIU, Ph.D. thesis, Northwestern Polytechnical University, 2001.
12. H. HU, in *Proceedings of the Metallurgical Reatyses*, edited by T. K. Tien and J. F. Eliot, (The Metallurgical Society, AIME, 1981) p. 385.
13. J. W. GIBBS *Trans Conn. Acad.* **III** (1876) 108 (1878) 343. Also in: "The Collected Works of J. W. Gibbs," Vol. 1 (Longmans, Green and Co., New York, 1928) p. 55.
14. J. WEISSMÜLLER, *Nanostruct. Mater.* **3** (1993) 261.
15. R. KIRCHHEIM, *Acta Mater.* **50** (2002) 413.

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