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Recrystallization in a powder metallurgy nickel-base superalloy

L. WINBERG
Department of Physics, Chalmers University of Technology, S-402 20 Gothenburg, Sweden

M. DAHLÉN
Department of Engineering Metals, Chalmers University of Technology, S-402 20 Gothenburg, Sweden

In an extruded powder metallurgy superalloy, carbide particles are precipitated at the previous particle boundaries. The role of these particles in the recrystallization behaviour has been studied by electron microscopy and hardness measurements. It was found that these carbide particles act as nucleation sites for recrystallization and inhibit growth of new grains.

1. Introduction

Nickel-based superalloys are an important class of high strength materials which effectively maintain their mechanical and corrosion properties at elevated temperatures. These alloys are most frequently used in the hottest parts of gas turbines and in hot corrosive environments in the chemical industry. Their excellent properties at elevated temperatures derive in the main from a combination of a high solute content, a large volume fraction of γ'-precipitation and a high chromium content [1].

The high level of alloying elements necessary to achieve the desired properties in superalloys often gives rise to segregation problems during casting. Fig. 1 shows the microstructure of a typical cast and wrought (CW) superalloy which exhibits segregation. The presence of areas lean in γ'-precipitation, the most important contributor to strength, reduces the mechanical strength of the material. By letting the melt solidify in the form of small powder particles, the segregation problems inherent in conventional casting and hot working of superalloys can be overcome. Powder metallurgy (PM) processing has been shown to be capable of substantially improving the strength and ductility of these alloys at low temperatures [2]. The prime reason for this improvement is that segregation is restricted to the size scale of each single powder particle. Microsegregation on this scale is easily removed by the heat treatment connected with further processing. At low temperatures the small grain size, which is sometimes inherited from the fast cooling of the powder particles, gives rise to high strengths, but at high temperatures this will tend to increase the ease of grain boundary sliding and thus reduce the creep resistance. Sometimes the grain size may be small enough to produce extended ductility bordering on superplasticity [3, 4]. If such materials are to be used in extreme high temperature service such as the hottest parts of gas turbines, it is of importance to improve their high temperature

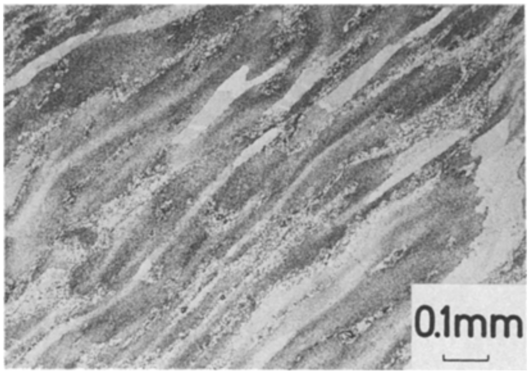

Figure 1 Structure of CW Astroloy showing rich and lean regions of γ'-precipitation.

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mechanical properties. One step towards this is to optimize the grain size by controlled recrystallization. Thus we have considered it of importance to investigate the parameters influencing the recrystallization behaviour of these powder metallurgical materials.

Carbide particles which are intentionally precipitated at grain boundaries in nickel-base superalloys impede grain boundary sliding and thus reduce the overall creep rate. The most frequent types of carbides are MC and $M_{23}C_6$. MC carbides are the most stable. They are nucleated at temperatures just below the alloy's melting temperature. In contrast $M_{23}C_6$ carbides are precipitated at lower temperatures (normally in the range 700 to 1000° C). In PM materials carbides precipitate at the surfaces of the powder particles during processing of the powder [5]. This results in a special carbide distribution in the densified material.

In many alloy systems precipitated particles such as carbides have been found to act as the nucleation sites for recrystallization. A special carbide distribution such as the one present in PM superalloys may very likely affect the recrystallization behaviour of these alloys. The present investigation is an attempt to determine the role of carbides and of their distribution in the recrystallization of PM superalloys.

2. Experimental

The commercial powder material which has been used in this investigation was produced by argon atomization of the nickel-base superalloy Astroloy. Its composition is given in Table I. The powder was consolidated by extrusion and Fig. 2 shows the structure obtained after a two hour heat treatment at 1150° C. This treatment was carried out prior to the experiments discussed here in order to dissolve the coarse γ'-phase, which otherwise could influence the recrystallization behaviour. The original powder particles, which had been elongated by the extrusion process, can be recognized because of the carbides which demarcate the previous particle boundaries (PPB).

Plane-parallel 10 mm cubic specimens of the material were deformed in compression between plane platens. The degree of deformation was 10% in all specimens. Since the nucleation sites of recrystallized grains can only be found as long as the fraction of recrystallized material is very small, very short recrystallization times were studied. Fig. 3 gives our definition of recrystallization time and shows a typical heating and cooling curve for the recrystallization treatments. Table II shows the

TABLE I Composition of Astroloy (wt%)

C	Cr	Co	Mo	Al	Ti	B	Ni	O
0.07	15.4	16.7	5.3	4.0	3.5	0.022	Bal	0.0098

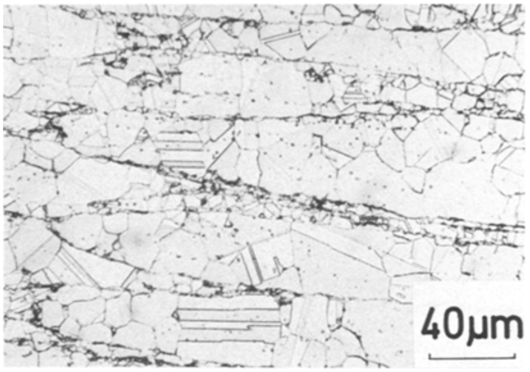

Figure 2 Microstructure of extruded Astroloy heat treated 2h at 1150° C. Note the elongated powder particles hedged-in by carbide particles.

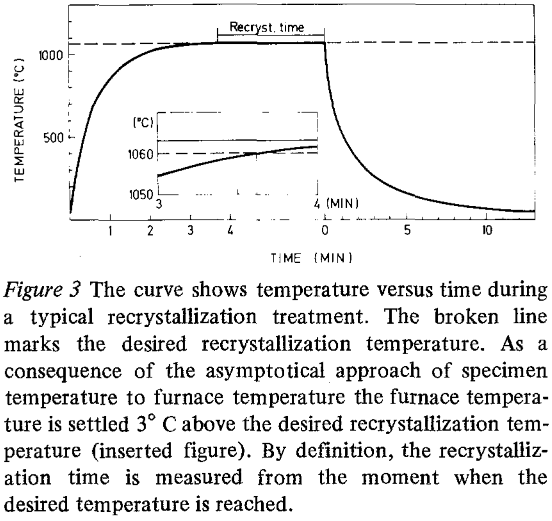

Figure 3 The curve shows temperature versus time during a typical recrystallization treatment. The broken line marks the desired recrystallization temperature. As a consequence of the asymptotical approach of specimen temperature to furnace temperature the furnace temperature is settled 3° C above the desired recrystallization temperature (inserted figure). By definition, the recrystallization time is measured from the moment when the desired temperature is reached.

TABLE II Times and temperatures used in the recrystallization treatments. The figures in the table mark the estimated percentage of recrystallized material

Temperature (°C)	Recrystallization time (min)				
	0	1	3	10	120
1020	$\bullet_{0}$				
1040	$\bullet_{0}$				$\bullet_{10}$
1060	$\bullet_{1}$	$\bullet_{1}$	$\bullet_{1}$	$\bullet_{10}$	
1080	$\bullet_{10}$				
1100	$\bullet_{10}$	$\bullet_{10}$	$\bullet_{10}$	$\bullet_{10}$	
1110	$\bullet_{30}$				
1130	$\bullet_{100}$				

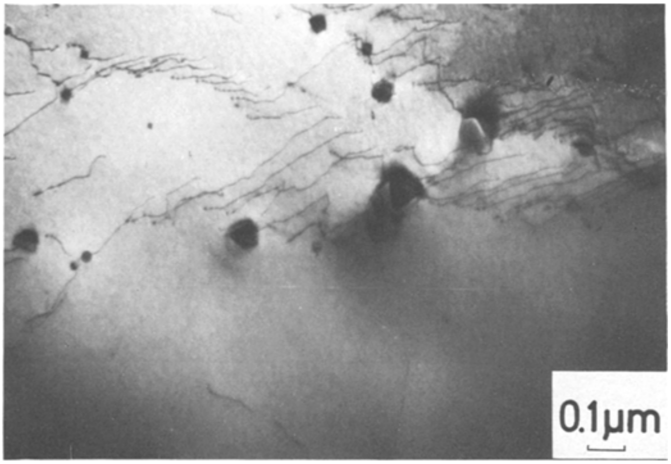

Figure 4 Electron micrograph of the material as undeformed. The particles are TiC situated in the PPBs.

recrystallization temperatures and times which were investigated in the experiments. In this table, zero recrystallization time is taken as the moment when the specimen has reached the specified recrystallization temperature. After the recrystallization treatments, hardness measurements were performed, followed by transmission electron microscopy.

3. Results

3.1. Microstructure

An electron micrograph of the extruded PM material prior to compressive deformation is shown in Fig. 4. The particles visible in the figure were identified by selected area diffraction as titanium carbide (TiC) and they were found to be preferentially located in the PPBs. The microstructure after 10% compressive deformation is shown in Fig. 5 where the dislocation arrangement around a carbide particle can be seen. Increased dislocation density in the vicinity of the particles compared to the dislocation density in particle-free areas was observed consistently in a number of different micrographs. On heating to temperatures of around 1000° C, the dislocation substructure changed, resulting in an even greater difference in dislocation density between areas close to the particle and in the particle-free areas (Fig. 6).

Fig. 2 shows that the material after consolidation and 2 h heat treatment at 1150° C had a duplex grain structure. In the carbide-rich PPB-regions the grains were small, because the carbides pinned the grain boundaries, while the carbide-lean areas contained much coarser grains. Electron microscopy verified that the coarse γ'-particles had been dissolved by the solution treatment at 1150° C. However, some relatively fine γ'-precipitation was present; this is ascribed to precipitation during quenching from the solution treatment temperature.

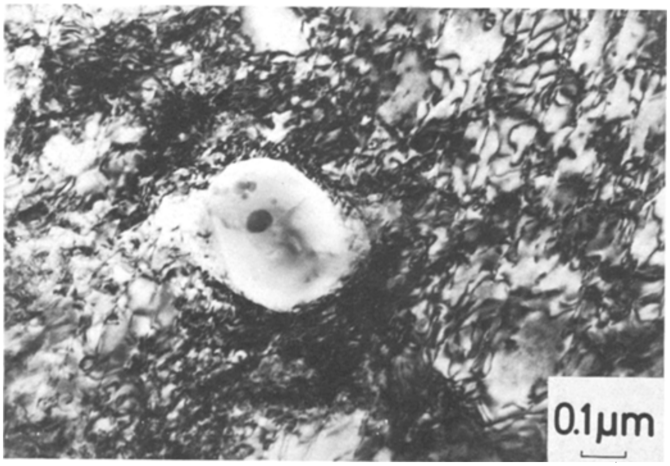

Figure 5 Electron micrograph of the dislocation structure around a carbide particle after 10% deformation.

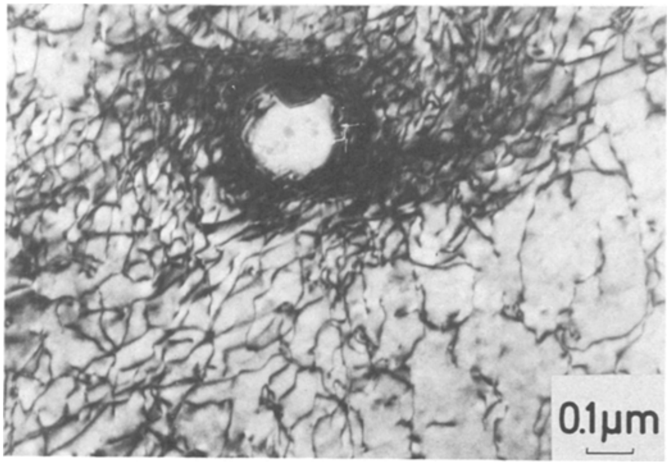

Figure 6 Electron micrograph of the dislocation structure after recovery at 1000° C for 1 h. Note the increased dislocation density gradient compared to Fig. 5.

Figure 7 Electron micrograph of a newly recrystallized grain associated with a carbide particle. The specimen was heat treated at 1060° C for 1 min.

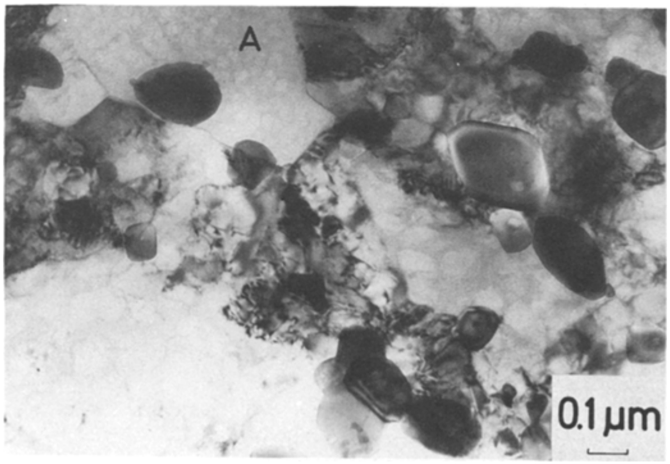

Figure 8 Electron micrograph of a recrystallized grain (A), surrounded by un-recrystallized grains (the light contrast of some grains is due to the diffraction conditions), in a carbide rich area. Note the carbide pinning of the grains. The specimen was heat treated at 1060° C for 1 min.

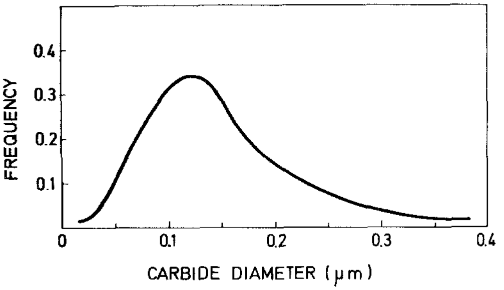

Figure 9 Carbide particle size distribution of the examined material.

3.2. Nucleation of recrystallization

A search was made by TEM to identify the nucleation sites of recrystallization. This revealed consistent association between newly nucleated grains and carbide particles (Fig. 7 and 8). The vast majority of new grains were in contact with one or more carbide particle. Only very few recrystallized grains seemed to be related to previous grain boundaries in positions where there were no visible carbides. The size distribution of the observed carbide particles is shown in Fig. 9. A special study was made of recrystallized grains which were connected with only one observable carbide. These carbides showed observable diameters in the range 0.15 to 0.30 μm. After correction for errors which arise when thin sections are used in particle size measurements [6] the true carbide diameters were calculated to be in the range 0.2 to 0.4 μm.

3.3. Growth of new grains

Grains in the carbide-rich regions were pinned by surrounding carbide particles that inhibited their growth. Fig. 8 shows the pinning of the interface between an old grain and a new grain (A) in a carbide-rich area of the specimen. The recrystallized grains were identified by means of γ'-particle distribution, which is different in new grains from that in unrecrystallized material; also the dislocation density is considerably smaller. The γ'-particle dispersions in both recrystallized and unrecrystallized areas are shown in Fig. 10.

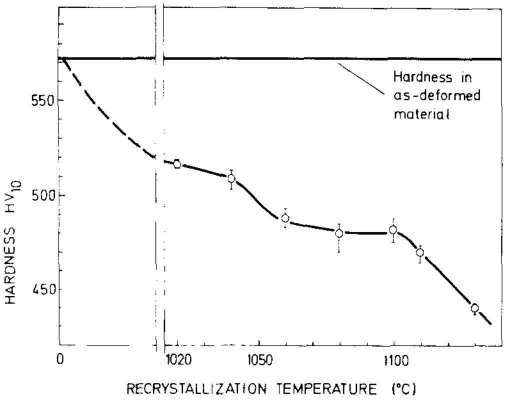

Figure 11 Hardness as a function of recrystallization temperature at recrystallization time 0. Note the plateau shape of the curve.

3.4. Isochronal recrystallization

Investigation at "zero" recrystallization time (i.e. after heating to the specified recrystallization temperature and immediate cooling) showed no sign of recrystallization for temperatures below 1060° C. At this temperature the degree of recrystallization was estimated to be about 1%. At a temperature of 1080° C the degree of recrystallization was estimated to be 10% and at 1100° C it was still 10%. However, at 1130° C the material was completely recrystallized within "zero" time.

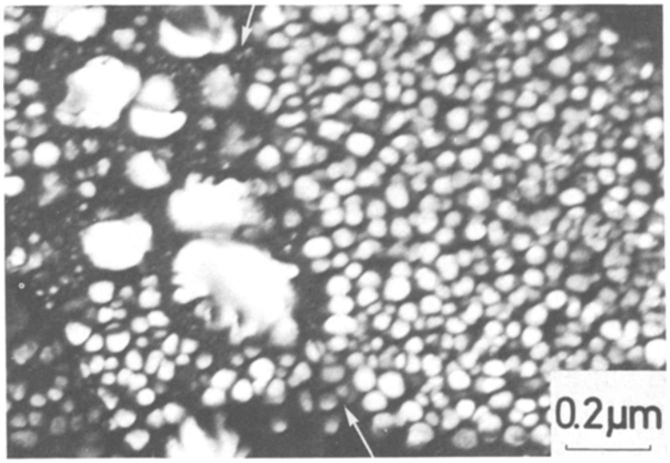

Figure 10 Dark-field electron micrograph of γ'-particle distribution in a newly recrystallized grain (right part) and in an un-recrystallized grain (left part). The arrows mark the low-angle grain boundary. The recrystallized grain shows a uniform γ'-particle size. The un-recrystallized part shows three different γ'-particle sizes. The large γ'-particles were coarsened during the recrystallization treatment, whereas the others were formed during the subsequent cooling. The finest size was formed in the denuded zones surrounding the coarsest γ'-particles. The specimen was heat treated at 1100° C for 3 min.

Fig. 11 shows the results of measurements of hardness versus recrystallization temperature at "zero" recrystallization time.

3.5. Isothermal recrystallization

Isothermal recrystallization was studied at 1060 and 1100° C for the times shown in Table II. At 1060° C the degree of recrystallization increased from an estimated 1% at zero time to 10% at 10 min. At 1100° C the degree of recrystallization appeared to remain stationary at 10% throughout the time period studied. In each case the recrystallized areas were situated at PPBs. Prolonged annealing at 1040° C, where no sign of recrystallization had been found at "zero" time, showed an estimated 10% recrystallization after 2 h.

4. Discussion

4.1. Nucleation of recrystallization

The observations of partially recrystallized material showed that in virtually every case the nucleation of recrystallization was connected with the presence of carbides. The very few examples where newly recrystallized grains were not connected with a visible carbide could be discounted due to the use of thin transmission specimens [6, 7], assuming that the carbide particle responsible for nucleation was not included in the thin foil. Carbide particles are thus considered to be the sole nucleation sites of recrystallization. According to the literature there should be a minimum critical size for second phase particles to act as effective nucleation sites. For example Nes [8] has suggested that in general this particle size should be around 0.5 to 1.0 μm. In some copper alloys [9] the critical particle size has been shown to be 0.3 to 0.5 μm, while in a medium carbon steel [7] a value of 0.45 μm was obtained. Our results of 0.2 to 0.4 μm are in good agreement with these previous values obtained in other alloy systems. In the present case, the critical particle size is slightly lower. This can be understood qualitatively when the special structure of the nickel-base superalloys is recalled. In most materials, small particles (< 0.3 μm) will be inactive as nucleation sites because they do not collect a sufficient density of dislocations around them. When edge dislocations reach a small particle they can easily bow around the particle and pass it, leaving behind prismatic loops. However, larger particles show no sign of prismatic loops adjacent to the particle. In this case dislocations appear to be attached to the particle as can be seen in Figs. 7 and 8. Superalloys always contain a dense precipitation of fine γ'-particles on a scale much finer than the dispersion of carbides. These particles are sheared rather than bypassed by the Orowan mechanism [10]. In consequence, dislocations do not move singly and independently but in groups associated by stacking faults of three basic types – superlattice intrinsic (or extrinsic) faults, antiphase boundaries and complex faults [11]. This, in effect, increases the friction stress that has to be overcome for dislocation movement, which in turn would tend to decrease the critical size at which carbides will be surrounded by a high enough dislocation density to become effective sites of grain nucleation.

4.2. Growth of new grains

Studies of both isochronal and isothermal recrystallization showed that after nucleation the subsequent growth of new grains was heavily retarded and that only the neighbourhood of the PPBs recrystallized at temperatures below 1130° C. There are two possible explanations of this behaviour, based either upon the distribution of dislocations or on the interparticle spacings.

During heating to the recrystallization temperature, recovery of the dislocation substructure occurs in areas of low particle density, while there is an increase of dislocation density in areas of high particle density. It has been shown [12] that recovery in this way increased the rate of nucleation but decreased the rate of growth of new grains. This mechanism in the present case would produce preferential nucleation in particle-rich areas, leading to full recrystallization only after annealing at higher temperatures.

The second explanation is as follows. In the PPBs the density of carbide particles is often very high, as can be seen in Figs. 2, 4 and 8. In these areas the interparticle spacing is of the order 0.5 μm. It has been shown [13, 14] that interparticle spacings of this order greatly decrease the rate of grain growth. The reason for this is that only a very few particles act as nuclei for recrystallization and the migrating grain boundaries have to move past "non-active" particles, which can act as pinning points for the migrating interphase.

From the above discussion an explanation can also be given for the step-wise hardness loss shown in Fig. 11. The curve shows three temperature intervals where the hardness drops rapidly, separated by two plateaus. The horizontal line indicates the hardness level of the as-deformed state. The first drop together with the subsequent plateau is ascribed to the lowering of dislocation density in the areas of low carbide density, since no sign of recrystallization was detected in this temperature interval. The next hardness decrease and plateau in the temperature interval 1040 to 1100° C is identified as recrystallization in areas of high carbide particle density. The degree of recrystallization in this temperature range was estimated to be 10% or less. Since recrystallization occurs in the areas of the highest dislocation density the hardness decrease can be expected to be considerable despite the small volume of recrystallized material. The growth limitation of the new grains discussed above explains the hardness plateau. The final slope of the hardness curve corresponds to the spreading of recrystallization into the interior of the previous powder particles, until the whole volume is fully recrystallized.

It was observed that the distribution of γ'-particle sizes in newly recrystallized grains was considerably different from that in unrecrystallized grains (Fig. 10). The coarse γ'-particles seen in unrecrystallized grains are considered to derive from precipitation during rapid cooling from the previous solution treatment at 1150° C and subsequent growth during heating to the recrystallization temperature. During air cooling the finer type of particles is precipitated. No coarse γ'-particles were observed in the recrystallized grains, indicating that the γ'-particles were dissolved in connection with the process of recrystallization. Cooling from the recrystallization anneal precipilated γ'-particles of a size similar to the finer population in un-recrystallized grains. Prolonged recrystallization treatment and subsequent cooling showed a further growth of the coarse γ'-particles in unrecrystallized grains, while there was no observable change in the γ'-particle size in recrystallized grains. This indicates that the γ'-particles in recrystallized grains are formed during cooling of the specimen from the recrystallization temperature. The explanation of γ' dissolution during recrystallization is as follows. The normal γ' solvus for the alloy occurs at 1130° C [15]. Recrystallizing grains tend to grow into grains with a relatively high misorientation. The γ' precipitate which is a coherent order phase must be re-formed in an orientation coinciding with the lattice of the new grain in which it finds itself after passage of the grain boundary. Otherwise, their surface energy would be drastically increased, which together with their high surface-to-volume ratio would make them extremely unstable. Thus in effect, the passage of a grain boundary into a γ' particle must lower its solvus temperature. When the grain boundary has passed, there remains a zone of supersaturation from which a precipitate of correct orientation will be formed. It is suggested that in view of the low degree of supersaturation at the recrystallization temperature, most of the reprecipitation occurs during air cooling. This would produce a similar dispersion to the fine part of the dispersion in un-recrystallized material.

5. Conclusions

(1) It has been shown that during primary recrystallization after straining the new grains are nucleated at MC-carbide particles. These carbides may be as small as 0.2 μm, which is slightly below the critical size established for other alloy systems. The difference is ascribed to the increase of the effective friction stress of the matrix by the fine γ'-precipitate distribution.

(2) The materials show duplex recrystallization behaviour. Areas corresponding to the previous powder particle boundaries which are rich in carbide particles, recrystallize at lower temperatures than the carbide-lean areas in the interior of the previous powder particles. This is because of the increased dislocation density surrounding carbide particles and also because of grain boundary pinning by these carbides.

(3) Existing γ'-particles were found to dissolve at the migrating interface between recrystallized and un-recrystallized grains. The re-precipitation of these particles is thought to occur not at the recrystallization temperature but during subsequent cooling.

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