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Inhibition of recrystallization in supersaturated solid solutions by large amounts of cold work

ERHARD HORNBOGEN
Institut für Werkstoffe, Ruhr-Universität Bochum, D-4630 Bochum, Germany

Recrystallization of a precipitation hardening γ-Fe-alloy was investigated as a function of defect density introduced by cold rolling. At $25\% < \epsilon < 90\%$ recrystallization took place in combination with precipitation, at $\epsilon > 90\%$ only particle growth controlled subgrain growth occurred. The results were explained on the basis of competition between two forces: an increased dislocation density producing an increased driving force for recrystallization and an increased rate of heterogeneous nucleation leading to the individual formation of particles which produce a retarding force.

1. Introduction

The rate of recrystallization of pure metals and solid solutions increases with increasing density of defects ρ_0, obtained for example by increasing the amount of cold work. This is due to the fact that formation and motion of recrystallization fronts (i.e. grain boundaries) become easier for higher defect densities. Temperature dependence at the start or at a certain stage of recrystallization t_R can be described phenomenologically as

$$t_{RO} = t_{RO}(\rho) \exp\left(+\frac{Q_R(\rho)}{RT}\right) \qquad (1)$$

where T is temperature, and the apparent activation energy Q_R, and even more the pre-exponential factor t_{RO}, decrease with the density of defects. The driving force for recrystallization F_ρ is proportional to the difference in defect density $\rho_0 - \rho_1$

$$F_\rho = \alpha\mu b^2 (\rho_0 - \rho_1) \qquad (2)$$

where α is a dimensionless factor that depends on the distribution of dislocations, μ shear modulus, b the Burgers vector, and ρ_0, ρ_1 the dislocation density after deformation (0) and after recrystallization (1).

There seems to be no reason why the rate of crystallization should not increase with the density of defects. Nevertheless it has been observed in supersaturated solid solutions [1, 2] and in alloys containing a dispersion of very small particles [3] that recrystallization is slowed down or even inhibited if the defect density is increased. In one case [3], this can be attributed to enhanced formation of recrystallization nuclei, if deformation is microscopically inhomogeneous at small and intermediate amounts of deformation because of cutting of thin particles. Many supersaturated solutions in which recrystallization is initiated at intermediate amounts of cold work cease to recrystallize at large amounts (Fig. 1). This effect has not only to be considered for soft-annealing

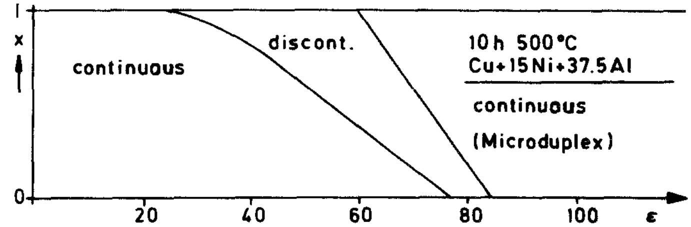

Figure 1 Portions of microstructural components x in a CuNiAl alloy heat treated 10 h at 500° C after different amounts of cold work – Recrystallization occurs in the field designated "discontinuous" [1].

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supersaturated solid solutions, but can also be used to produce microstructures which combine a high defect density, fine dispersion of particles, and a pronounced texture (see Fig. 6). As an example the annealing behaviour of a precipitation hardening γ-iron alloy will be described as a function of the amount of cold work, and a general explanation for the conditions under which the observed anomaly occurs is attempted.

TABLE I Composition of the alloy (Balance is Fe).

Element	wt%	at.%
Ni	28	27
Cr	16	15.5
Mn	1.5	1.5
Al	0.34	0.70
Ti	2.35	4.65
Mo	1.40	0.81
V	0.43	0.53
Si	0.50	0.98
C	0.09	0.40
B	0.01	0.06
P	0.01	0.03
S	0.01	0.02

2. Experimental procedure

The composition of the alloy is given in Table I. This alloy was quenched from 1100° C to obtain a homogeneous solid solution which was subsequently deformed by rolling between 0 and 98.6% reduction in thickness and isothermally annealed in the temperature range between 600 and 900° C. The details of the precipitation behaviour have been described earlier [4]. The metastable γ'-phase forms coherently, and is replaced by the stable incoherent η-phase which forms exclusively by heterogeneous nucleation. Recrystallization was studied by light- and transmission electron microscopy and by texture analysis with X-rays using the {1 1 1} reflections.

3. Experimental results

As usual, the time before the start of recrystallization t_R decreases after small amounts of cold work, however the trend is reversed at about 30% cold work, while above 90% no recrystallization is found (Fig. 2). Quantitative measurements of the portions of the microstructural components for one heat treatment (t, T = const) are shown in Fig. 3. Only in the range designated "discontinuous" is recrystallization found. Small amounts of cold work are insufficient to induce recrystallization and amounts larger than 90% cause disappearence of recrystallization. Typical microstructures are shown in Figs. 4 and 5. In Fig. 4a recrystallization proceeds into the deformed material in which the metastable γ'-phase is precipitated. Recrystallization occurs in combination

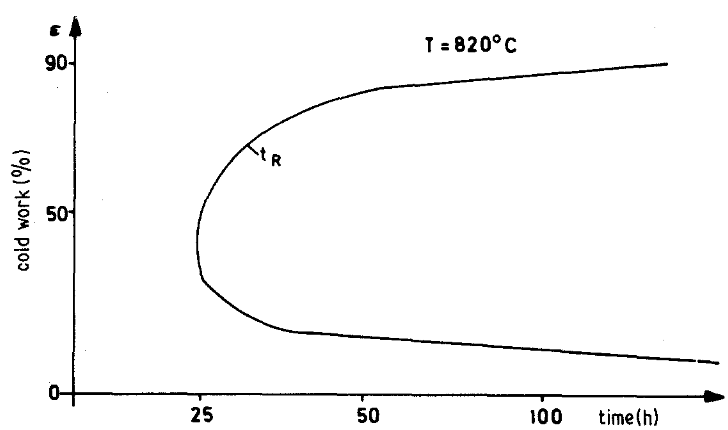


Figure 2 Start of recrystallization as a function of amount of cold work and period of ageing at 820° C (composition see Table I).

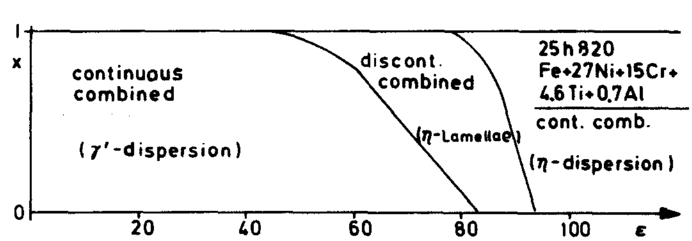


Figure 3 Portion of microstructural components for one heat treatment and different amounts of cold work, recrystallization in the field designated "discontinuous combined".

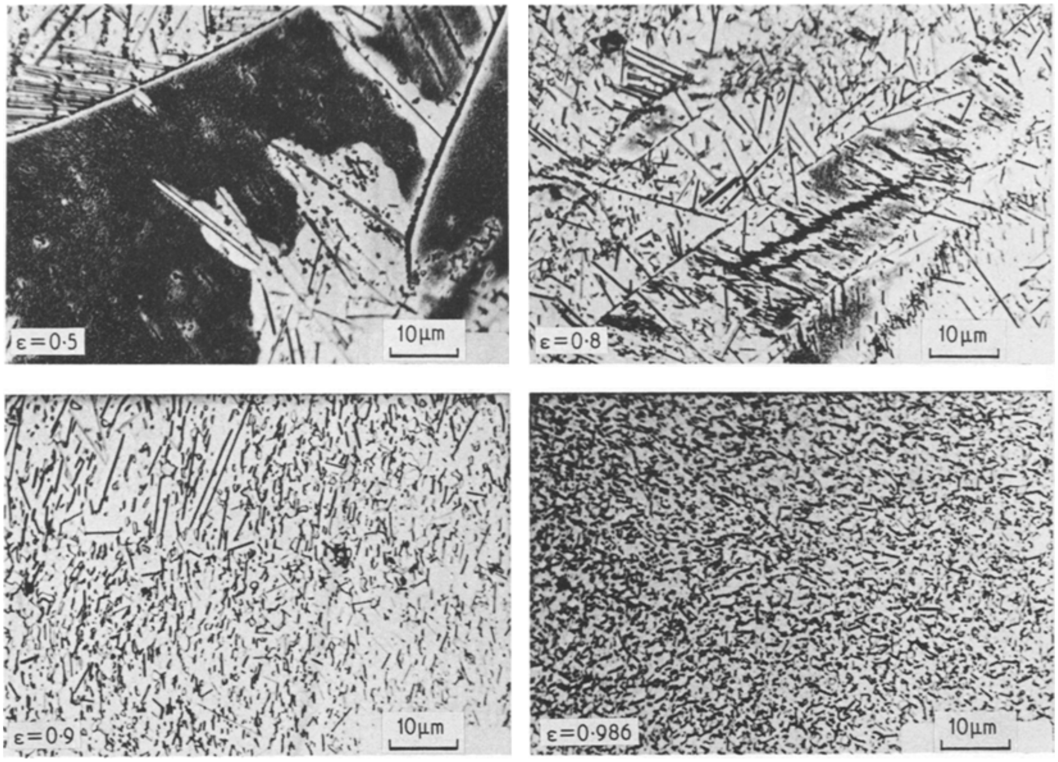


Figure 4 Microstructures after a heat treatment of 25 h at 820° C preceded by different amount of cold work ϵ, (light-microscopy).

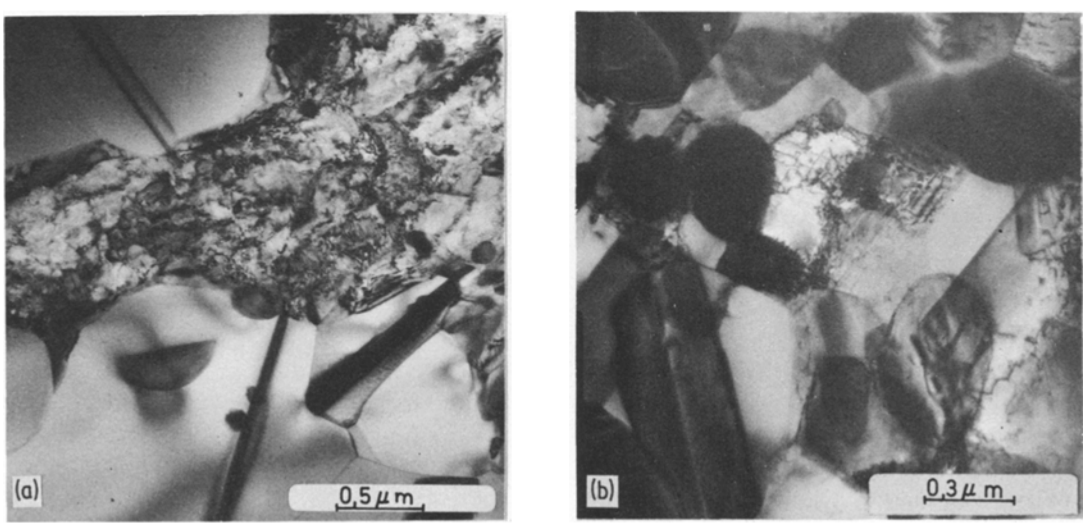


Figure 5 (a) partially recrystallized conditions (combined discontinuous reaction) 50 h, 750° C, $\epsilon = 90\%$ (b) *in situ* recrystallized condition (combined continuous reaction) 264 h, 750° C, $\epsilon = 98.6\%$ (transmission electron microscopy).

with the precipitation of the stable η-phase, which forms as plates. In Fig. 4b recrystallization is almost complete (see Fig. 7b). In figure 4c few plates (which indicate that recrystallization had occurred) are left, and in Fig. 4d the microstructure is characterized by a dispersion of almost spherical particles. Additional TEM investigations indicated that the particles were incoherent and that no reorientation by the motion of high angle boundaries had occurred. Subgrain growth only took place at $\epsilon > 90\%$ (Fig. 5). This implies that the texture should be preserved by the heat-

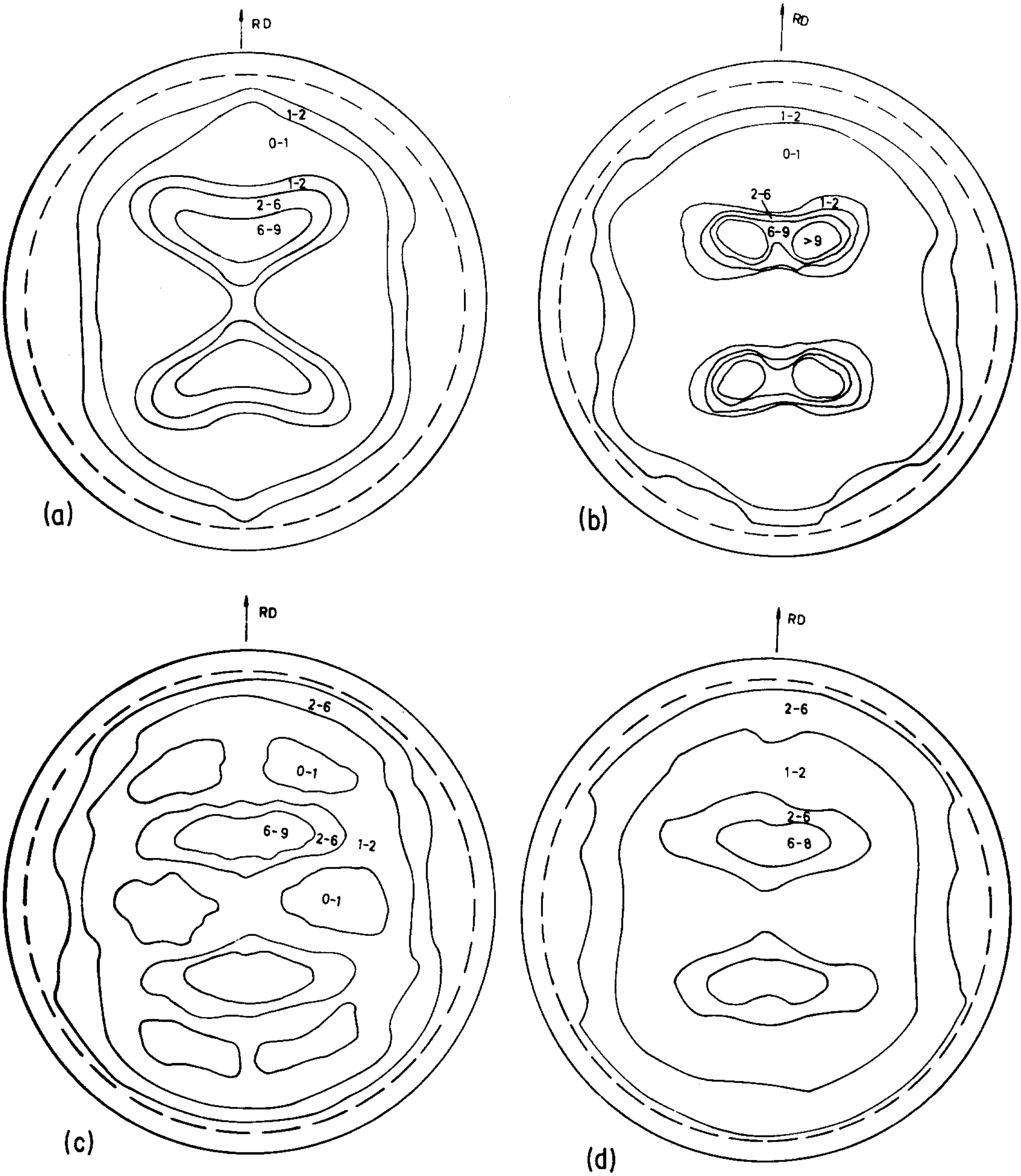
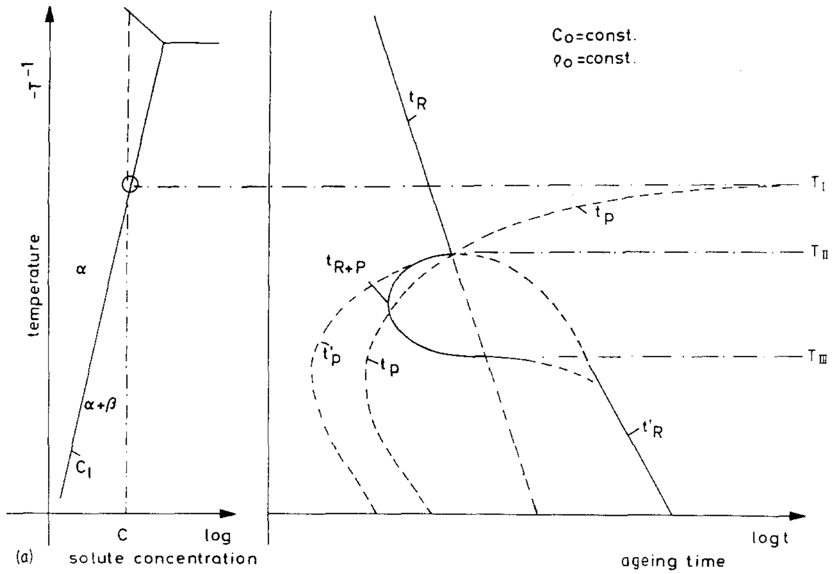


Figure 6 {1 1 1} pole figures after different thermomechanical treatments (1 – 2 random orientation): (a) 98.6%, as rolled (b) $\epsilon = 98.6\%$, 50 h at 820° C (c) $\epsilon = 50\%$, 50 h at 820° C (d) $\epsilon = 50\%$, 100 h at 820° C

treatment which led to the microstructure shown in Fig. 4d. In Fig. 6 the pole figures of specimens that were annealed after intermediate and large amounts of cold work are compared with the rolling texture. It is evident that the rolling texture appears even more pronounced after annealing the highly deformed alloy. The intermediate amount of cold work (50%) yielded metallographic evidence for about 70% recrystallization. This is in agreement with a drastic increase in randomness of the orientation and a decrease of the intensity of the original rolling texture.

4. Discussion

The anomalous dependence of recrystallization rate on the defect density can be interpreted, if it is considered that forces additional to the driving force F_ρ (Equation 2) will act at a recrystallization



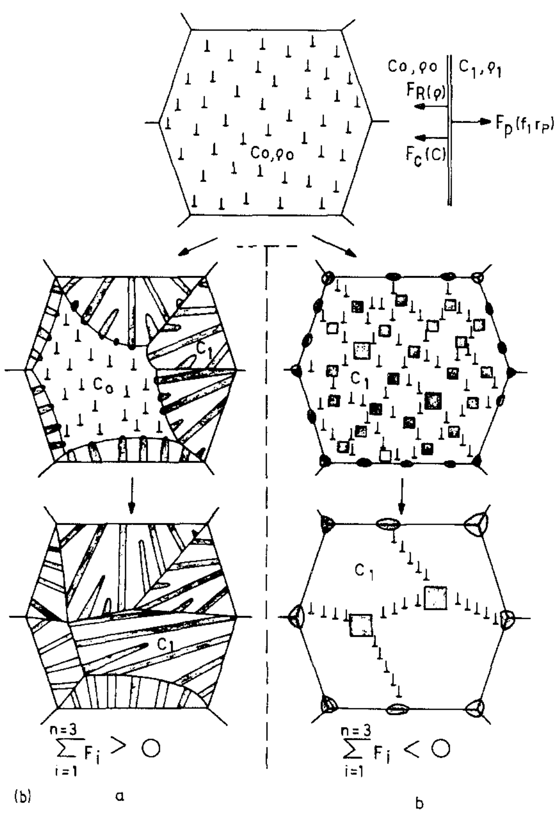


Figure 7 (a) Schematic representation of temperature dependence for start of recrystallization t_R and precipitation t_P. $t_R + t_P$ combined recrystallization + precipitation, t'_R recrystallization retarded by precipitated particles. (b) development of microstructures from a defect super-saturated solid solutions discontinuous (a), continuous (b).

front. Forces due to segregation will be neglected, only alloys in which precipitation can occur are discussed. The temperature dependence of their crystallization behaviour is shown in Fig. 7a. It has to be considered that the temperature dependence for the start of precipitation t_P has a different *C*-shape from that of recrystallization (Equation 3). It is highly dependent on the particular defect structure, if a phase exists in equilibrium that forms preferredly by heterogeneous nucleation at certain lattice defects.

A survey can be obtained for the temperature dependence of the different reactions from Equation 1 for the start of recrystallization and Equation 3 for precipitation

$$t_P = t_{PO} \exp\left([\Delta G_N(T,\rho) + Q_D]/RT\right) \qquad (3)$$
$$t_P \lesseqgtr t_R$$

t_R and t_P are the times before recrystallization or precipitation start at a temperature T. The activation energy for nucleation ΔG_N decreases with undercooling and the possibility of heterogeneous nucleation at defects. The activation energies for diffusion Q_D, like Q_R, (Equation 1) are regarded as temperature independent. This leads to the different shape of the two functions $t_R = f(T)$ and $t_P = f(T)$ from which four temperature ranges can be defined (Fig. 7):

$T > T_E = T_I$	recrystallization in the solid solution
$T_I > T > T_{II}$	recrystallization precedes precipitation
$T_{II} > T > T_{III}$	recrystallization and precipitation occur simultaneously
$T_{III} > T$	precipitation retards crystallization

$T_I = T_E$ is the equilibrium temperature of the particular alloy composition. The temperatures T_{II} and T_{III} depend on defect density, composition, and especially on the nucleation behaviour of the defect alloy.

Increasing the defect density or decreasing the solute concentration leads to a decrease of T_{II}, so that for such conditions recrystallization may always precede precipitation (range II).

Range III is characterized by the fact that with increased supersaturation, precipitation and recrystallization occur simultaneously by a new reaction which will be designated as "combined dis-discontinuous reaction" (Fig. 7b). In range IV precipitation occurs by individual heterogeneous nucleation and so rapidly that the pinning force of the particles effectively inhibits the motion of any recrystallization front. The only reaction that can occur is particle-controlled subgrain growth which is termed "combined continuous reaction" (Fig. 7b). The rate of the discontinuous combined reaction and the transition to the continuous mechanism can be derived by quantitatively evaluating and summing the forces F_i, that act as the reaction front [2]:

$$\text{(range III)} \sum_i^{n=3} F_i = F_R + F_C - F_P > 0 \quad (4)$$

$$\text{(range IV)} \sum_i^{n=3} F_i = F_R + F_C - F_P < 0 \quad (5)$$

The forces are defined as the change in Free Energy per unit area A across the reaction front, that moves in x-direction:

$$F = \frac{1}{A} \frac{dG}{dx}$$

The driving force F_ρ has been discussed already. The change in concentration $c_0 - c_1$ gives rise to a chemical driving force.

$$F_C \approx \frac{RT}{V_m} c_0 \ln \frac{c_0}{c_1}. \quad (6)$$

(R = gas constant, V_m = molar volume). Opposed to these driving forces is the pinning force, which exists if a random dispersion of particles has formed ahead of the reaction front:

$$-F_P = \frac{3 f \gamma_{\alpha\alpha}}{2 r_P}, \quad (7)$$

f is the volume fraction, r_P the radius of the particles and $\gamma_{\alpha\alpha}$ the energy of the grain boundary. There are several possibilities for grain boundary particle interactions including blanking-off discs from grain boundaries (from which the formula for F_P was derived), dissolution and re-precipitation, transformation of metastable → stable, and dragging.

The discontinuous combined reaction can lead to accelerated recrystallization and the greatest possible rate of approach to equilibrium at a given temperature. It usually produces a relatively coarse lamellar aggregate of phases (Figs. 4a and 5a).

A mixed microstructure of dislocations arranged to sub-boundaries and a fine dispersion of particles is formed by the continuous reaction (Figs. 4d and 5b). Here equilibrium is approached at the slowest possible rate.

It follows from Equations 4 and 5 that the occurrence of the continuous or the discontinuous reaction is not only a function of the temperature and time but also of the defect density. If the dislocation density is increased by increasing amounts of cold work, not only the driving force F_R is increased but also the force $-F_P$, provided that the dislocations or defects that form by dislocations provide sites of easy heterogeneous nucleation of the equilibrium phase. In addition the concentration c_0, and therefore the available chemical driving force F_C, is reduced. This provides the basis for an explanation for inhibition of recrystallization at high defect densities. In Figs. 8a to c typical examples are shown schematically for the sequence in which the particular forces F_ρ, F_c, F_P can change during an isothermal ageing sequence. In Fig. 8a recrystallization is complete, before precipitation has started, and before the retarding force F_P could act. In Fig. 8b, precipitation and recrystallization occur simultaneously and both driving forces are additive and disappear. If, due to enhanced heterogeneous nucleation, precipitation has started before recrystallization, the retarding force can become larger than the driving force and no motion of recrystallization fronts is possible. If the forces (for a certain temperature and time of annealing) are shown as a function of defect density in the alloy, a critical defect density ρ_c for the condition

$$(F_c + F_\rho) = F_P \quad (8)$$

can be defined. For inhibitation of recrystallization (which had been initiated by smaller amounts of cold work) it is necessary that above a certain defect density ρ_N nucleation becomes so rapid

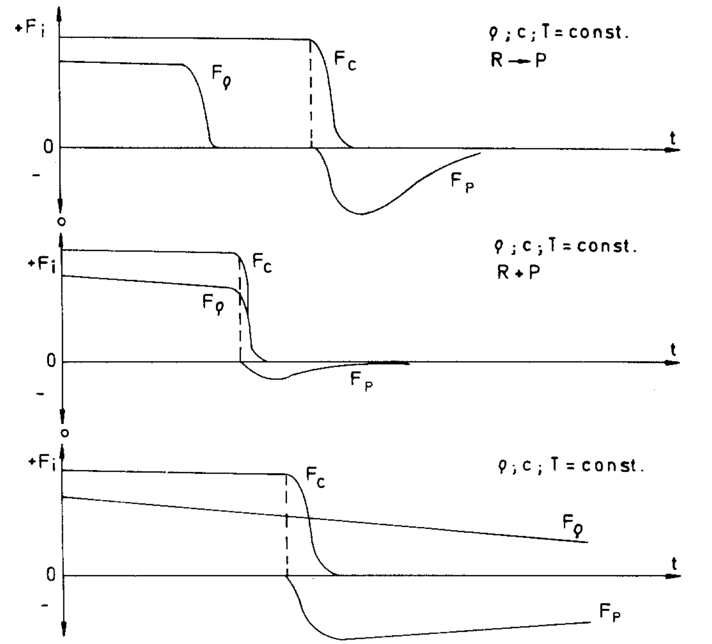


Figure 8 Driving and retarding forces during isothermal ageing sequences: ρ, C, T = constant: (a) recrystallization and precipitation occur simultaneously (c) precipitation inhibits recrystallization.

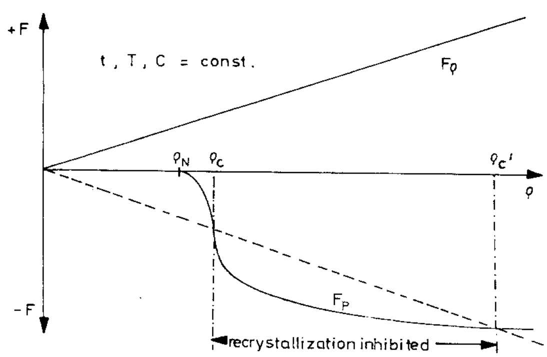


Figure 9 Driving and retarding forces as a function of defect, density (or amount of cold work) c, T, t = constant. ρ_N dislocation density above which the stable phase can be nucleated, ρ_C dislocation density above which driving force F_ρ is overcompensated by retarding force F_P.

that there is a fine dispersion of particles and consequently a high retarding force F_P originates. The necessary condition for this transition to occur is

$$\frac{dF_P}{d\rho}(\rho_c - \rho_N) \geqslant \frac{dF_\rho}{d\rho}\rho_c + F_c \qquad (9)$$

This condition is demonstrated in Fig. 9 for the simplified case that the chemical driving force $F_c = 0$ due to completion of precipitation.

General predictions for the alloy systems in which the transition from recrystallization to inhibition of recrystallization is expected, can be based on Equation 9. The requirement of a relatively high dislocation density $\rho > \rho_N$ for the formation of the equilibrium phase implies that structures caused by a reaction of several dislocations such as sub-boundary nodes or segments with grain boundary-like structure are the catalytic sites for nucleation rather than the individual dislocation lines. Only if the formation of such sites becomes copious due to a high initial defect density, a sufficiently high retarding force F_P can arise and in turn inhibit the formation and motion of recrystallization fronts. For alloys in which the individual dislocations are suitable to nucleate the equilibrium phase, the critical dislocation density for inhibition ρ_c (Fig. 9) can become so small that no recrystallization will occur at all. As Fig. 9 indicates there should exist a very high defect density $\rho_{c'}$ at which the driving F_ρ again surpasses the retarding force F_p. However, in many cases this is not observed, because the defect density which can be produced in the usual ways, such as cold rolling or wire drawing, is not sufficient to overcompensate the retarding force of the precipitating particles.

5. Summary

The recrystallization behaviour of a precipitation hardening γ-Fe-alloy was investigated as a function of defect density as produced by cold rolling. The alloy recrystallizes at intermediate amounts of cold work $25\% < \epsilon < 90\%$ by a reaction in which recrystallization is combined with precipitation of the equilibrium phase. Above 90% cold work, recrystallization is completely inhibited and only subgrain growth takes place. The discontinuous process induces random orientation of the crystallites while the very pronounced rolling texture is preserved by the continuous reaction.

The general conditions under which this phenomenon is expected are as follows:

(1) A homogeneous supersaturated solid solution into which defects have been introduced.

(2) Nucleation of the equilibrium phase is heterogeneous, but individual dislocation lines are insufficient for rapid nucleation of the equilibrium phase.

(3) Therefore the nuclei become copious and evenly dispersed only above a critical defect density ρ_N which permits the formation of enough sites for heterogeneous nucleation to produce a fine dispersion of particles.

(4) The condition for inhibition of recrystallization is that the retarding force caused by the particles F_P above ρ_N increases so much more rapidly than the driving force F_p with increasing defect density, that the condition $F_P > F_\rho$ (Equation 5) is fulfilled.

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