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Annealing of Supersaturated and Deformed Al-0.042 wt % Fe Solid Solutions

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The recrystallisation and precipitation behaviour of a quenched Al–Fe alloy (0.042 wt % Fe) during isothermal annealing was observed, after different amounts of plastic deformation, by microscopy and measurement of electrical conductivity. Three temperature ranges with different types of reaction can be distinguished with decreasing temperature: (i) The recrystallisation of solid solution; (ii) A two-step reaction: the recrystallisation is complete before precipitation starts; (iii) One-step reactions: characterised by a mutual influence of recrystallisation and precipitation.

The combined reactions can take place by two different mechanisms. These are *continuous recrystallisation*, by growth of sub-boundaries, with simultaneous precipitation and coarsening of particles, and *discontinuous recrystallisation*, by motion of high-angle boundaries dissolving dislocations and redistributing particles. Conclusions of practical annealing treatments are drawn from the knowledge of the micromechanisms.

1. Introduction

Frequently metastable alloys which contain foreign atoms in supersaturation as well as a non-equilibrium concentration of lattice defects have to be heat-treated. To acquire equilibrium atoms and defects have to anneal out by precipitation, recrystallisation and recovery respectively. Heat-treatments of such alloys have to be conducted to achieve completely different final microstructures. For instance a microstructure can be required which contains dislocation networks and a fine dispersion of particles, in order to get an additive hardening effect, due to these two types of obstacles to motion of dislocations. Often, on the other hand, a heat-treatment is required which leads, under the most economical conditions, to the softest possible state of the alloy.

In order to obtain these microstructures under optimum conditions, the reactions which occur in defect-containing and supersaturated alloys have to be understood. The basis for the understanding is the different temperature-dependence of the rate of annealing of lattice defects (in our case the dislocations) and of atoms, as given in fig. 1 and in earlier papers [1, 2].

From fig. 1 it follows that for such alloys three basic temperature ranges can be expected:

$T > T_E$: normal recrystallisation in solid solution;

$T_E > T > T_{RP}$: recrystallisation complete before precipitation has started;

$T < T_{RP}$: simultaneous healing out of dislocations and precipitation.

(T_E: equilibrium temperature for the solid solution, T_{RP}: temperature at which recrystallisation and precipitation start at the same time.)

It is well known that the two processes have a reciprocal effect on each other. An alloy con-

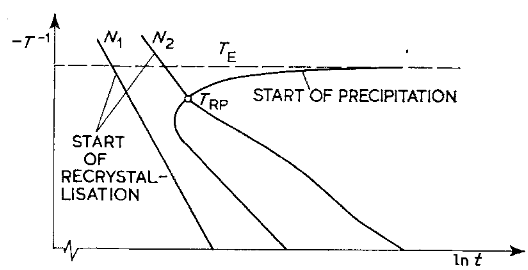

Figure 1 Schematic time-temperature-reaction diagram for two different dislocation densities ($N_1 > N_2$), assuming that only the recrystallisation and not the precipitation is influenced by dislocations.

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taining lattice defects has a nucleation behaviour and precipitation kinetics different from a perfect crystal. In a supersaturated solid solution recrystallisation is affected in such a manner that the motion of a reaction front can either be impeded, if particles have already precipitated in front of it, or accelerated, if precipitation occurs simultaneously with the healing out of dislocations at the reaction front.

This situation can be characterised quantitatively by the introduction of forces of different signs acting at the reaction fronts [1]. A special situation may occur in such alloys if the forces which retard the motion of grain-boundaries are larger than the driving forces. Under this condition no reaction front can develop, and the defects have to heal out by a continuous process. The dislocations rearrange to sub-boundaries, and with increasing time the diameter and the angle of misorientation of the subgrains will grow.

The purpose of this investigation is to study the different types of recrystallisation which occur if an alloy with a certain density of defects and a certain concentration of atoms in supersaturation is aged at different temperatures. Besides the practical importance of Al-Fe alloys, this system seemed to be suitable for a basic investigation on these phenomena because of the following reasons:

(i) Previous investigations indicate that iron precipitates from aluminium without the formation of metastable phases [3]. Therefore a relatively simple precipitation behaviour can be expected.

(ii) The lattice defects produced by plastic deformation of aluminium are all undissociated dislocations, so that these are the only type of defects which have to anneal out to reach equilibrium.

Therefore this system seemed to offer a simple example for an attempt to examine how far the schematic diagram (fig. 1) represents the situation occurring in real supersaturated and defect alloys.

In order to obtain a homogeneous solid solution an Al-0.042 wt % Fe alloy was chosen. The maximum solubility is 0.052 wt % (0.026 at. %) at $T = 655°$ C. The iron precipitates as Al_3Fe with monoclinic structure [3]. Previous investigations [4, 5] showed that the activation energy for diffusion of Fe in Al is extremely high. In addition a strong influence of even very small amounts of iron on the recrystallisation has been reported [3, 6, 7]. In the present investigation it is attempted to give an interpretation of this phenomenon based on microscopic mechanisms.

2. Experimental Procedure

The experimental method is a combination of measurements of electrical conductivity and microscopy. The first method is a very sensitive probe for the kinetics of annealing out of even extremely small amounts of dissolved atoms [8] as well as for the annealing out of defects [9]. Because both effects lead to changes of the conductivity in the same direction, this macroscopic method had to be supplemented by microscopic investigations.

It is known that for the study of recrystallisation processes a combination of light and electron microscopy is necessary in order to give a full description of the phenomena. The interaction of precipitating atoms with defects left in the crystal can only be made visible by electron microscopy, while light microscopy allows one to survey the size and distribution of recrystallised grains.

Details concerning the measurement of electrical conductivity used in this investigation can be obtained elsewhere [10]. The samples for the electron microscope were polished by a jet technique, and for the light microscope they were etched by anodic oxidation [11]. After homogenisation at 645° C the samples were deformed by cold-rolling and then isothermally annealed at various temperatures between 150 and 500° C in a salt bath.

3. Experimental Results

3.1. Micromechanism of Precipitation

As found in previous investigations [4], iron precipitates from the undeformed alloy as semicoherent flat needles (in $\langle 100 \rangle$ direction and with $\{110\}$ habit) by homogeneous nucleation in the matrix (fig. 2a). In addition, grain-boundaries act as sites for heterogeneous nucleation of particles of irregular shape with almost incoherent interfaces. Nucleation at individual dislocations was not observed.

Furthermore the micromechanism of precipitation was observed in alloys which had been deformed at room temperature by 10, 50, and 90%. The micrographs (figs. 2b, c) show the typical morphologies and nucleation sites of the particles. It was not possible to detect a difference in crystal structure as compared to those precipitating in the undeformed alloys. However, the morphology of the particles changes. Fig. 2b shows precipitates in a 50% deformed alloy. The dislocations have rearranged into regular networks and preferred particle nucleation is found at special dislocation nodes in networks with relatively small mesh size. Nucleation at the normal $a/2\ \langle 1\,1\,0\rangle$ components of the network is again not observed.

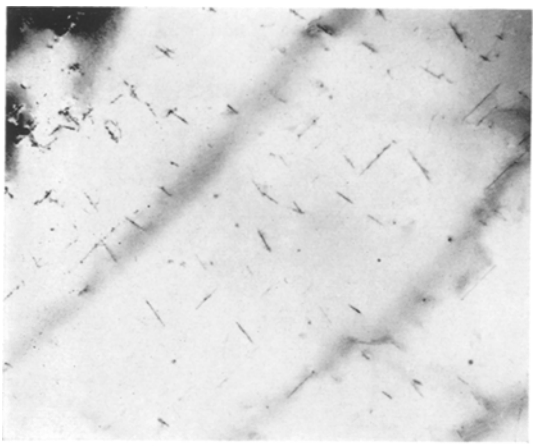
The morphology of the particles changes to approximately spherical shape if the misorientation between the subgrains is increased by high amounts of cold-work and subsequently rearranging the new dislocations into subboundaries. This indicates a transition from semi-coherence when particles form in a perfect lattice (fig. 2a) or at separate dislocations, to an almost isotropic interface if they form at high-angle boundaries (fig. 2c).

The temperature-dependence of the start of precipitation in differently deformed alloys is shown in fig. 5; the derivation of this figure is explained later.

3.2. Healing out of Dislocations

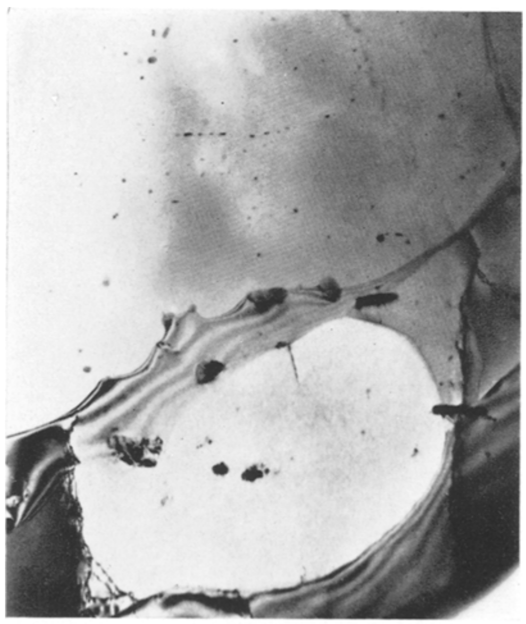
In the deformed alloy dislocations begin to redistribute themselves even at room temperature, so that a 90% deformed sample, for example, already contains subgrains of about 0.1 μm diameter.

The further healing out of dislocations can occur, basically, by two mechanisms [12]. The micrographs (fig. 3) show that in some areas the formation of crystallites of new orientations is visible, while in other areas no changes of the microstructure can be resolved. The investigation by transmission electron microscopy (figs. 2b, 2c, 3d) indicates that in the last-mentioned



(a)

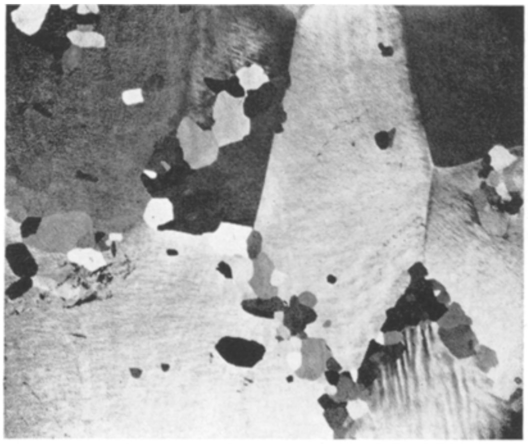
(b)



(c)

Figure 2 Electron micrographs of precipitates in differently deformed alloys. (a) Undeformed, homogeneous nucleation, 6000 h 300° C (×7500); (b) 50% deformed, plates precipitating at dislocation networks, 7500 h 250° C (×13500); (c) 90% deformed, spheres precipitating at sub-boundaries, 5 min 410° C (× 20500).

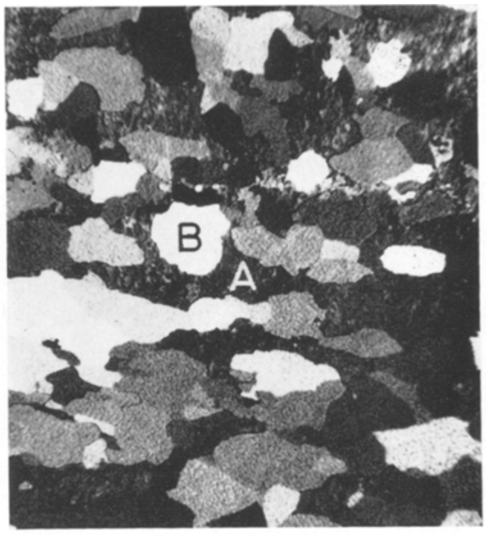
areas a continuous process occurs which is characterised by growth of the subgrains which are present at the beginning of the heat-treatment with a simultaneous increase of their angle of



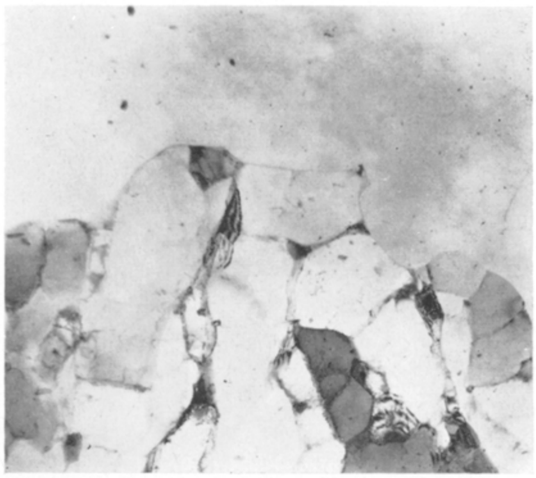
(a)

(b)

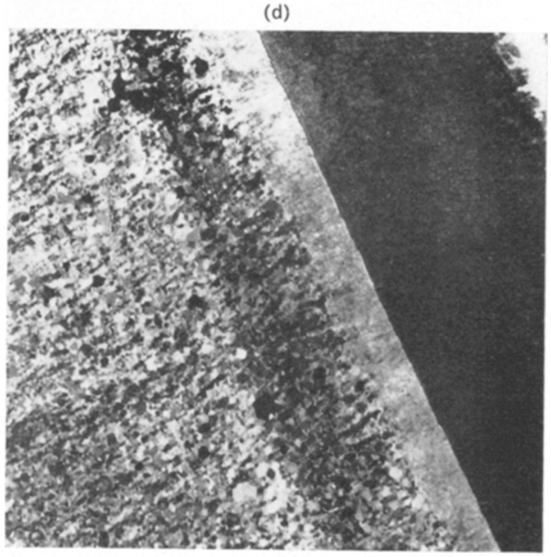
Figure 3 Start of recrystallisation. (a) At grain-boundaries, 50% deformed, 10 sec 500° C (× 23); (b) at deformation bands, 90% deformed, 20 min 350°C (×54); (c) in the grain where continuously recrystallised parts (A) are dissolved by discontinuous reaction fronts (B), 90% deformed, 118 h 250° C (× 83); (d) electron micrograph of a reaction front similar to (c), 50% deformed, 7500 h 250° C (× 5900); (e) partly continuously recrystallised alloy, 90% deformed, 10 min 350° C (× 56). (a, b, c, e) light micrographs, etched by anodic oxidation [11].



(c)



(d)



(e)

misorientation. If the specimen is aged for a sufficiently long time, these subgrains can reach a size which can be resolved in the light microscope (fig. 3e). This condition is then termed as "continuously recrystallised".

It follows from figs. 2b, c that the rearrangement of the sub-boundaries is affected by precipitation. In this connection two situations can be distinguished:

(i) The dislocations rearrange before nucleation has started. In this condition iron atoms will be segregated at the dislocations which form the sub-boundaries.

(ii) After nucleation has started these nuclei will act as sinks for iron atoms which are segregated at certain sites of networks, and during their growth the concentration of segregated atoms will decrease.

Depending on the structure of the sub-boundaries at the beginning of nucleation, the density of nuclei at these sub-boundaries can be quite different. When they contain a small density of particles, dislocations can rearrange themselves easily after segregation zones have been removed by nucleation. The particles which had been present at such boundaries are still visible in fig. 2c. Boundaries which were able to offer nucleation sites in high density will now be so pinned by particles that they become immobile according to the Zener condition [10], and at the same time further growth of the particles will occur. These boundaries will only be able to rearrange themselves after the pinning force of the particles has been reduced due to their coarsening. This first process we refer to as *continuous recrystallisation*.

In the second process the dislocations heal out by motion of a reaction front with the structure of a high-angle boundary. This process is normally known simply as recrystallisation. We distinguish it as *discontinuous recrystallisation*. It requires the formation of such a boundary and forces sufficient to move it through the deformed alloy.

The light micrographs, figs. 3a, b, c, give evidence for the sites at which this type of reaction starts in differently deformed alloys. At small amounts of deformation (50%) the original grain-boundaries are the preferred sites at which this type of reaction can start (fig. 3a). In 90% deformed alloys the process occurs, in addition, at the so-called deformation bands (fig. 3b). These are bands at which in the interior of the grains a large misorientation has been produced by plastic deformation. After annealing, condensation of dislocations at these sites will easily lead to the formation of high-angle boundaries which, subsequently, can start to move as reaction fronts.

If the conditions for formation and motion of these fronts are fulfilled, the two types of reaction, continuous and discontinuous recrystallisation, will always occur simultaneously (fig. 3c). The progress of continuous recrystallisation will reduce the driving force for discontinuous recrystallisation because of a decreased dislocation density.

If the subgrains are able to grow relatively big before they are dissolved by the discontinuous process, a microstructure can result, which consists of aggregates of grains, formed by discontinuous and by continuous recrystallisation. Such microstructures were observed frequently if the specimens had been aged at low temperatures (fig. 3d).

The motion of the reaction front has a small effect on the dispersion of the particles. They are influenced by grain-boundary diffusion so that accelerated precipitation or coarsening occurs (fig. 2c).

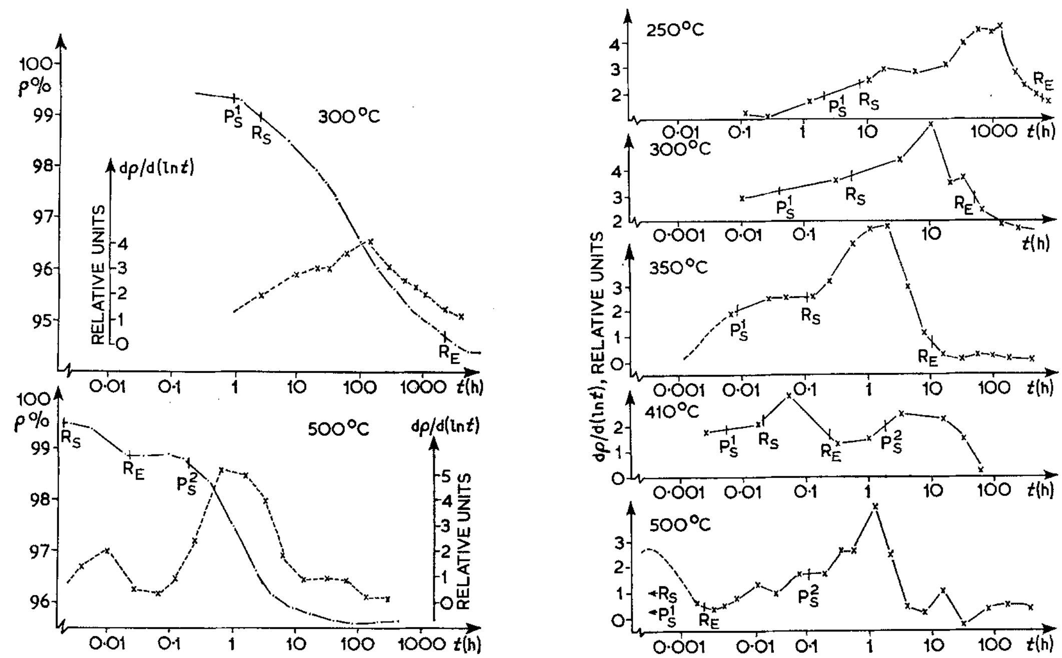
If the discontinuous process is complete before the composition of the matrix has reached its equilibrium value, after continued ageing homogeneous nucleation of particles, similar to fig. 2a occurs, in addition to the continued growth of the particles already present due to previous heterogeneous nucleation at defects. Exclusive homogeneous nucleation occurs if the discontinuous recrystallisation has been so rapid that no heterogeneous nucleation could start before the defects had annealed out. As expected, this is the case at higher annealing temperatures.

The analysis of microstructure and resistivity shows that at low temperatures (150° C) during the ageing period used in this investigation (10 000 h) neither precipitation nor a considerable rearrangement of dislocations had occurred.

3.3. Measurements of Electrical Conductivity

The basic shapes of the electrical resistivity (ρ) vs. time (t) curves obtained after isothermal ageing at different temperatures are shown in fig. 4a. It seemed practical to plot the differentiated curve $d\rho/d\ln t$, because the reaction steps become more evident (figs. 4a, b).

Fig. 4b indicates that for the 90% deformed alloy at low temperatures only one maximum is



(a) (b)

Figure 4 Resistivity ρ during isothermal ageing at different temperatures. (*a*) ρ(——) and dρ/dln$t$ (- - -) plotted vs. annealing time; 50% deformed alloys aged at 300° C (one-step reaction) and 500° C (two-step reaction); (*b*) dρ/dln$t$ vs. time plots for 90% deformed alloys; with increasing temperature the one-step reaction changes into the two-step reaction. R_S, R_E: start and end of recrystallisation; $P_S{}^1$ $P_S{}^2$: start of precipitation before and after complete recrystallisation.

measured, while with increasing temperature the separation into two maxima is found.

The correlation of these macroscopic results was made by observation with the light and electron microscopes. From this it became clear that at lower temperatures the process of healing out of dislocations and precipitation of iron atoms occurs simultaneously, so that only one maximum appears. At higher temperatures little or no iron can precipitate during the recrystallisation. Therefore a second maximum occurs after the recrystallisation has been completed, due to precipitation that starts by homogeneous nucleation in the recrystallised matrix. Under favourable conditions recrystallisation can be completely finished before any precipitation can start (fig. 4a, 500° C).

The times at which recrystallisation or precipitation have started according to the microscopical investigations, have been marked on the resistivity curves in figs. 4a, b.

3.4. Time-Temperature-Reaction Curves

The results obtained by the combination of the three experimental methods are shown in figs. 5a, b, c. It follows from these diagrams that the start of precipitation occurs earlier with increasing amount of deformation. However, the time at which the precipitation is completed is affected much less. Figs. 5b, c show that with increasing temperature more and more iron is left in the matrix after recrystallisation is completed. This gives rise to the second reaction step.

Under the conditions described in this paper the type of the reaction changes several times as a function of the annealing temperature. Therefore the curves cannot be used for the determination of activation energies, unless investigation of the microstructure has indicated that only one process takes place in a given temperature range. It is however preferable to determine data on diffusion of Fe in Al by reactions other than precipitation [4, 5].

4. Summary and Conclusions

The following conclusions can be drawn from the investigations of an Al-0.042 wt % Fe alloy:

(i) The interfacial structure and shape of the particles can vary from semicoherent and thin plates after homogeneous nucleation in the

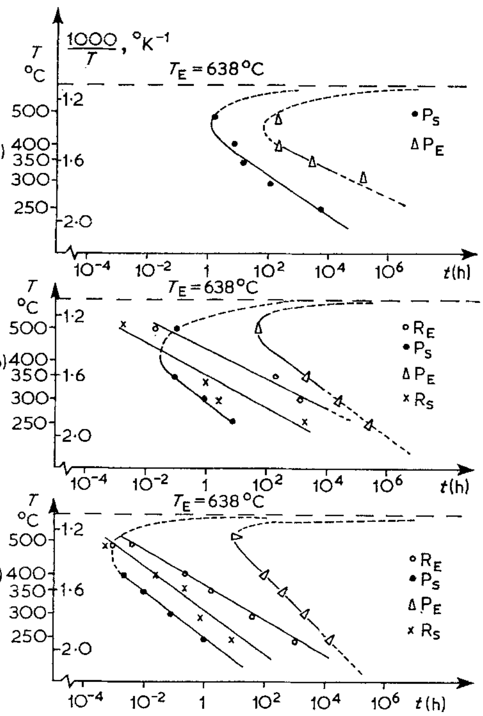


Figure 5 Time-temperature-reaction diagrams for the start and end of recrystallisation (R_S, R_E) and precipitation (P_S, P_E). Deformation: (a) 0%; (b) 50%; (c) 90%.

matrix to non-coherent spheres after heterogeneous nucleation at defects with grain-boundary structure.

(ii) The start of precipitation is accelerated owing to preferred nucleation at certain types of defects.

In all specimens precipitation could not be completed by growth of the particles formed at the heterogeneous nucleation sites. After recrystallisation was completed, homogeneous nucleation always occurred in the matrix, now free of defects, with the result that precipitation continued up to about the same time for all the alloys, irrespective of degree of deformation.

An additional factor that influenced the distribution of particles was discontinuous recrystallisation. The reaction front accelerated precipitation due to grain-boundary diffusion while it moved through the crystal. If an alloy with given composition and defect concentration was aged at different temperatures, three ranges always were observed:

Aged above T_E: recrystallisation only.

Aged below T_E and above T_{RP}: first healing out of dislocations, then precipitation.

Aged below T_{RP}: for the conditions given in these experiments particles precipitate and defects heal out simultaneously by two reactions – continuous and discontinuous recrystallisation – and as a consequence a mixed microstructure is observed.

The portion of the discontinuously recrystallised microstructure should increase with decreasing supersaturation and dislocation density. The case of pure continuous recrystallisation was not observed under the experimental conditions used.

The following conclusions for practical heat-treatments of these alloys can be deduced. The alloy can be soft-annealed most quickly in the temperature range of the two-step reaction when the recrystallisation is not impeded by precipitation. The start of the precipitation is delayed very much due to the high activation energy of diffusion of iron in aluminium. Therefore a microstructure can be obtained in deformed alloys containing dislocations populated by a large concentration of segregated iron atoms. In this state, the work-hardened condition can be preserved up to about 200° C for very long periods of time.

If a microstructure is desired which contains both dislocations and precipitates, the annealing conditions have to be chosen so that only continuous recrystallisation occurs. This means ageing at high supersaturations and low temperatures.

Two different types of annealing textures can be expected for this alloy; for the two-step process the normal annealing texture should be found as for the homogeneous solid solution, and for the alloy which has been entirely continuously recrystallised the rolling texture should be preserved. At medium temperatures when both kinds of recrystallisation occur, a mixture of these textures should be obtained by a macroscopic pole figure determination [1].

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