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Grain growth and secondary recrystallization in iron

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The effect of impurities and of slight deformations as factors modifying the kinetics of normal grain growth after primary recrystallization in pure iron is investigated. Samples of pure iron and Armco iron, each with or without a content of precipitated oxides, were examined.

Inclusions inhibit grain growth up to high temperatures, whilst the influence of strain depends on its amount: very low deformations (~2%) generally block grain growth; for deformations around 5%, secondary recrystallization takes place; higher deformations (~10%) accelerate the early stages of growth, leading to not very large final grain dimensions. 2.0% elongation has been found to be, in our conditions, the critical strain limit for secondary recrystallization.

1. Introduction

The motion of grain boundaries during primary recrystallization is due to the energy introduced into the metal by deformation; the amount of energy stored during plastic strain is to a great extent coincident with the energy of dislocations, whose density is raised in cold-worked metals by several orders of magnitude [1]. The grain growth which occurs after nucleation in primary recrystallization therefore consists of the migration of a boundary which moves through a highly deformed volume and transforms it into a relatively strain-free one, reducing its dislocation density to low values; the difference of free energy between the new strain-free grains and the highly-strained adjacent matrix is the driving force of the process [2–5].

After completion of primary recrystallization, the whole polycrystal is relatively strain-free, and the grains, now in contact with one another, have approximately the same low dislocation density. At suitably higher temperatures, a new grain-growth process sets in. In these conditions the main driving force of the growth is the surface energy of large-angle grain boundaries [5, 6]; the boundary moves because the increase of the mean size of the grains reduces the total surface of the interfaces in the overall polycrystal volume, and therefore reduces the total boundary energy. The free energy of this process is relatively slight [7]; the energy stored during cold-working is greater [1] by one or two orders of magnitude even without very large deformations. The great difference between the values of the two kinds of driving energies easily accounts for the fact that the boundary surface tension becomes rate-determining for boundary migration only when the strain introduced into the metal is very low; for high and medium deformations, the driving force due to cold-working energy is predominant.

The process by which grain size increases after primary recrystallization can occur according to two different patterns [6, 8]. (1) *Normal* (*or continuous*) *grain growth*, during which all the crystals grow in a uniform way; (2) *abnormal* (*or discontinuous*) *grain growth*, consisting in an exaggerated growth of only a few larger grains at the expense of the many smaller ones. This second kind of process, also known as *secondary recrystallization*, occurs in the presence of conditions which can restrain or inhibit normal grain growth and transform its kinetics.

Recent progress in preparation of highly pure metals and in knowledge of polygonization and textures has made it possible to attempt a more exact definition of the conditions under which secondary recrystallization can be observed: the present paper studies, from this standpoint, the effect of impurities and of slight deformations in pure iron. The work is mainly concerned with the kinetics of growth processes; it tries to define the different growth processes only from the standpoint of the kinetics of the changes occurring in mean linear sizes, without taking into consideration the statistical distribution of crystal dimensions during growth.

2. Materials

Different types of iron were used in the present research, to study the effect of different impurities on grain growth.

(a) High purity iron* prepared by us according to a method previously described [9, 10]; total metal impurities <20 ppm, (C + N) <3, 0 < 5 (S + P) <10.

(b) Oxidized high purity iron*, obtained from pure iron P by a thermal treatment in moist hydrogen to introduce a certain amount of precipitated oxide (0 ~ 600 ppm).

(c) Technically pure Armco iron*; total metallic impurities ~0.05%, (C + N) ~0.03%, 0 ~ 0.03%, (S + P) ~0.03%.

(d) Deoxidized Armco iron*, obtained by a thermal treatment at 1480°C in pure dry hydrogen, in order to eliminate O, C and N; total metallic impurities 0.05%, (C + N) <10 ppm, 0 < 10 ppm, (S + P) ~0.005%.

The characteristics of the different types of iron listed above may therefore be summed up as follows, from the standpoint of the different impurities: P, virtual absence of any type of impurities; PO, no interstitial and substitutional impurities; presence of precipitates; AD-no interstitials and precipitates; presence of substitutionals; A, presence of interstitials, substitutionals and precipitates.

3. Experimental methods

A first lot of specimens was obtained by cold-rolling (80 to 90% reduction in section) and subsequent annealing at 670°C for 3 h in pure hydrogen, thus obtaining completely recrystallized samples with a linear mean grain size of 30 to 70 µm (10 to 15 µm for Armco specimens), and 2 mm thickness.

To evaluate the effect of different amounts of cold-working, another series of specimens was prepared in a similar manner, with a final shape of square rods (section 7 × 7 mm^2). Afterwards, the specimens were deformed by tension (2 to 24%) cold-rolling (5 to 30%), and compression (4 to 39%)†.

The thickness of the specimens was chosen sufficiently high to avoid restraint of grain growth due to the sheet-thickness effect [11–13].

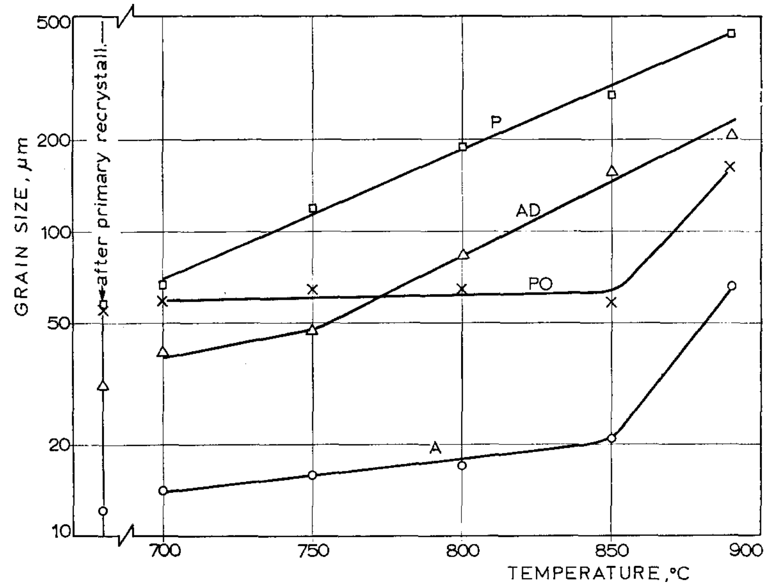

Figure 1 Grain growth after primary recrystallization for specimens of pure iron (P), oxidized pure iron (PO), Armco iron (A) and deoxidized Armco (AD). Isochronal anneals (3 h) at increasing temperatures.

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* Codes used in the present paper: P for pure iron, PO for oxidized pure iron, A for Armco iron, AD for deoxidized Armco iron.

† The amount of deformation is always given as per cent elongation or compression.

The thermal treatments (isochronal and isothermal) were performed in a purified argon atmosphere; the specimens were contained in a quartz tube immersed in a molten salt bath, kept at constant temperature, in order to reach the required temperature (regulated to ±1°C) as soon as possible.

After each anneal, micrographs were taken; the linear mean grain size was measured on each micrograph by means of the intercept method. X-ray and hardness measurements were also performed.

4. Effect of impurities and deformation

The normal grain growth in unstrained specimens was studied on the samples completely recrystallized, as described in the preceding paragraph. The variation of grain size was followed during successive isochronal anneals (3 h) at increasing temperatures, and the data obtained are shown in Fig. 1.

Two types of trends can be pointed out: a continuous regular increase of grain size is shown by the specimens with no precipitated impurities (curves P and AD), whilst the curves

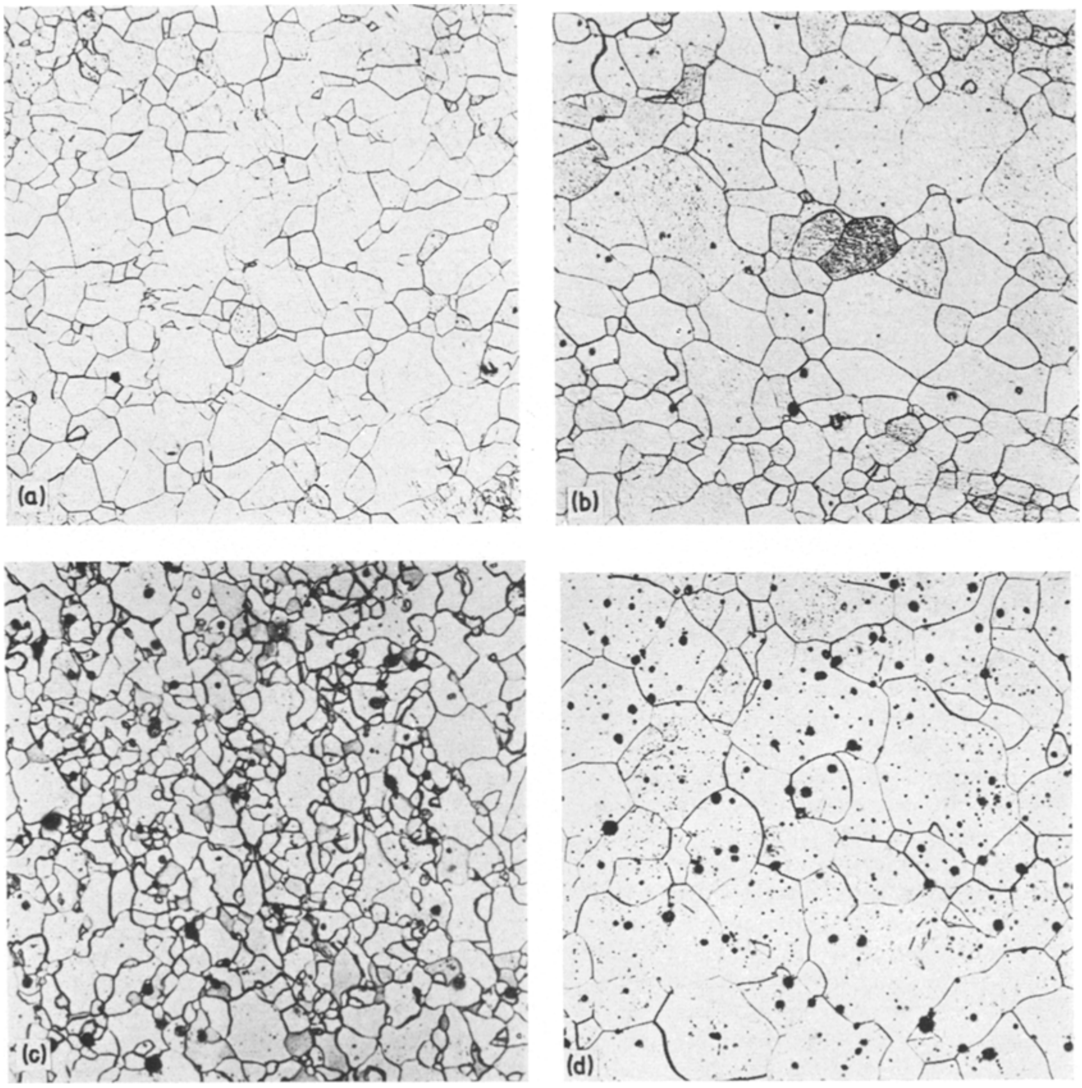


Figure 2 Unstrained pure iron (P) annealed at (a) 800°and (b) 850°C. Unstrained Armco iron (A) annealed at (c) 850° and (d) 890°C. Magnification: (a) ×18; (b)×18; (c) ×240; (d) ×120.

PO and A, pertaining to specimens containing an appreciable fraction of precipitated oxides, show practically no grain size increase up to temperatures as high as 850°C.

In Fig. 2, micrographic evidence of the two types of growth can be seen; in the specimen containing oxides, the grain-size distribution is very similar to that characteristic of discontinuous grain growth.

It is important to note that AD samples, which have relatively high contents in substitutional impurities but no oxide precipitates (since these were eliminated by high-temperature treatment in dry hydrogen) show nearly the same behaviour as the high-purity iron.

It would therefore appear that the main obstacle to grain growth is represented by precipitated particles while (as distinct from that which applies to primary recrystallization [10]) substitutional impurities play a less important role, and do not to any appreciable extent modify the shape of grain-growth curves: their effect is only a reduction of grain-size increase.

It is possible that the blocking effect of precipitated impurities is particularly effective for a random-type distribution of the inclusions, as in our case; it is not excluded that different types of distribution can have different effects, favouring a less homogeneous distribution of grain sizes. A statistical analysis of grain sizes and of the distances between the precipitated particles is in fact the object of a separate investigation.

The modification produced by slight deformations in the normal growth of recrystallized specimens were studied only on three kinds of samples (P, AD and A, the behaviour of PO and A being analogous); for each of these, three amounts of deformation were given; $\sim$5, 15 and 25% strain. The specimens, as said, were deformed at room temperature, by tension, rolling or compression.

The present paper essentially reports results obtained on the samples strained by tension; in some of the diagrams, however, since the difference between the various types of deformation are not relevant as far as the present discussion is concerned, the results pertaining to the three straining techniques were averaged together.

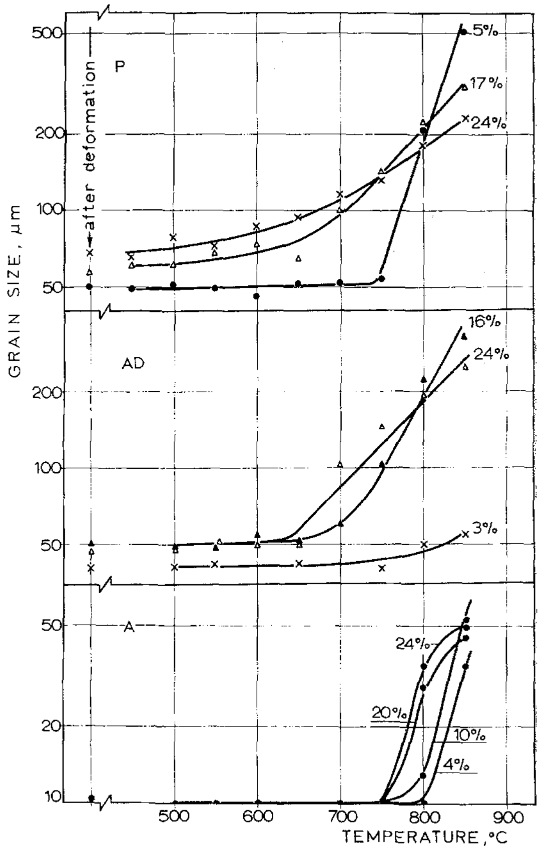


Figure 3 Grain growth for specimens strained in tension (the different amounts of deformation are indicated). Pure iron (P), Armco iron (A), deoxidized Armco iron (AD). Isochronal anneals (3 h) at increasing temperatures.

Fig. 3 shows the evolution of linear grain size during a series of 3 h anneals at increasing temperatures. Two stages of growth may be singled out: the first stage shows larger growth for more deformed specimens (in particular, the specimens with deformation around 5% do not show any growth at all). However, during the second stage, an inversion tends to occur; in particular, the materials cold-worked around 5% undergo a sudden growth, their mean grain size eventually becoming the largest. Only for the samples with the slightest deformations ($\sim$3%) did the growth continue to be blocked; the presence of polygonized dislocations (evidenced in the micrograph of Fig. 4) seems to be responsible for this behaviour. This pattern becomes particularly evident in the scheme of Fig. 5a.

The behaviour described, clearly evident for pure iron, is also observed in deoxidized Armco iron, thus confirming that substitutional impurities do not qualitatively affect grain-growth behaviour of specimens.

A sharp displacement towards higher temperatures must be remarked for Armco iron

Figure 4 Polygonized dislocations in Armco iron deoxidized (AD) strained 3 %, after anneal at 650° C (×660).

(containing inclusions of precipitated oxide), though the same trends outlined above seem to appear in this case too, shifted to a higher temperature range.

The dependence of grain growth on the rate of deformation is further analysed in the schemes of Figs. 5b and c. All the schemes of Fig. 5 average the results obtained on the specimens deformed by tension, rolling or compression.

Fig. 5b characterizes the deformation-dependence of secondary growth at 700° C (first stage), showing the ratio

$$\frac{\text{mean grain diameter at } 700^\circ\text{C}}{\text{mean grain diameter before annealing}}$$

as a function of the amount of cold-work; Fig. 5c gives an analogous plot calculated for 850° C (second stage). The results, at both stages, indicated the possibility of collecting the data into only two curves, the first one referring to both the specimens without oxide inclusions

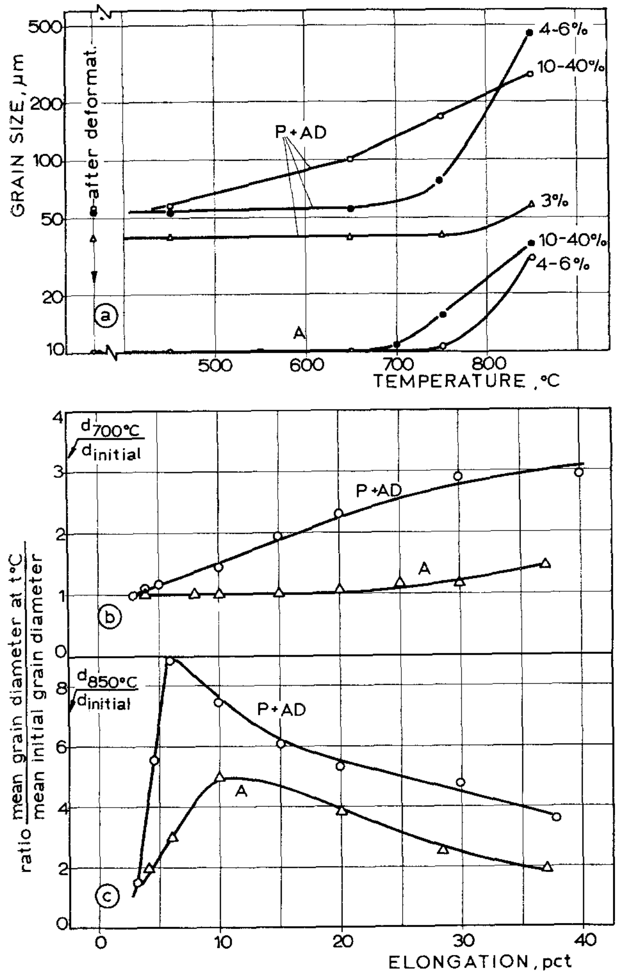


Figure 5 Schemes obtained by averaging data pertaining to specimens deformed by tension, rolling and compression. Pure iron (P), Armco iron (A), deoxidized Armco iron (AD). (a) grain growth dependence on temperature; isochronal anneals (3 h) at increasing temperatures; the ranges of strain are indicated. (b) grain growth dependence on the amount of deformation; relative growth at 700° C. (c) grain growth dependence on the amount of deformation; relative growth at 850° C.

(P + AD), the second one to the oxidized specimens (A). The above observations regarding Fig. 3 are here clearly evidenced: in the first stage of growth, higher deformation rates involve higher growth rates; in the second stage a peaked curve is obtained, and maximum growth rates are obtained for an intermediate amount of deformation (~5% for pure iron, 10% for oxidized iron); very low growth rates are observed for very slight cold-working (~3%), and, after the peak, increasing deformations (10 to 40%) produce decreasing growth.

The whole analysis of the results suggests a comparison between the effect of impurities (Fig. 1, unstrained specimens) with the effect of slight deformations (Fig. 3 and 5a, deformed specimens) on secondary growth. Precipitated impurities and small amounts of strain may be characterized as two conditions strongly modifying the trend of normal grain growth in metals after completion of primary recrystallization. Probably, the defects produced by very low strains and the inclusions present in the matrix both act as networks of pinning points: in the first case a lattice of polygonized dislocations, in the second a network of precipitated particles may inhibit grain-boundary motion, thus blocking grain growth up to high temperatures.

5. Kinetics of grain growth

The results discussed in the preceding section showed that it was possible to study both precipitates and polygonized dislocations as factors inhibiting normal grain growth in recrystallized metals.

Different authors [11, 13–15] have studied grain-growth kinetics, in the absence of any inhibiting factor. In these conditions, the mathematical formulation proposed by Feltham [15] forecasts that boundary motion should start immediately at zero time with a maximum rate, decreasing later with time according to a parabolic trend. In our case of slightly deformed specimens (as well as for materials containing precipitated oxides), the growth kinetics are modified by an initial stage in which grain-boundary motion-rate is nil or nearly nil, for a time depending on temperature.

The study of the kinetics of growth was performed by means of a series of isothermal anneals on two kinds of specimens, pure iron P and Armco iron A. The specimens were strained in tension by about 2, 5 or 10%.

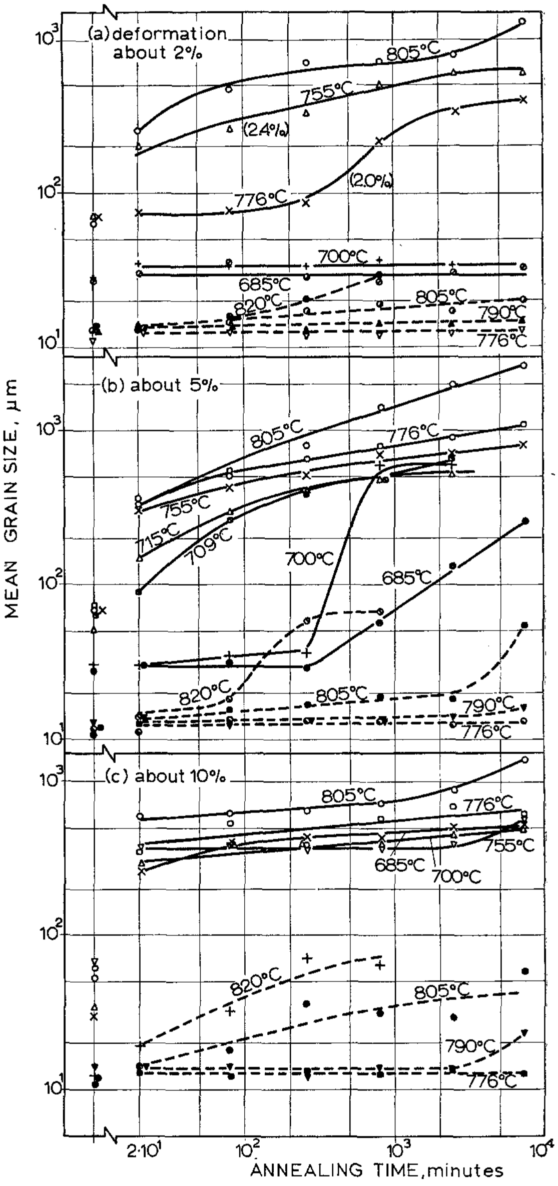


Figure 6 Isothermal grain growth for specimens strained in tension. Solid lines: pure iron (P). Dotted lines: Armco iron (A).

In Fig. 6 the isothermal secondary growth is plotted for different temperatures. Since it is possible to distinguish two types of trends, depending on the range of annealing temperatures, the results pertaining to pure iron are averaged in Fig. 7a for the lower temperatures (685 to 700° C) and in Fig. 7b for the higher ones (755 to 805 °C).

As regards the specimens strained about 5%, while at the highest temperatures it is possible

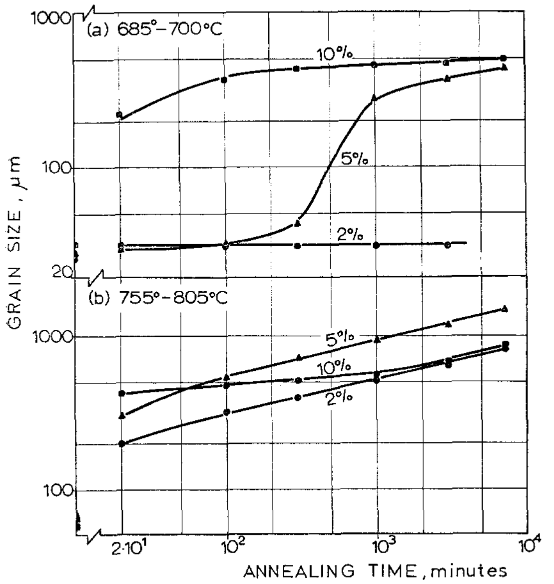


Figure 7 Isothermal grain growth for specimens strained in tension. Pure iron. The different amounts of strain are indicated. (a) scheme averaging results obtained at lower temperatures (685 to 700° C); (b) scheme averaging results obtained at higher temperatures (755 to 805° C).

to observe a sharp increase of mean grain size even after short annealing time, the data pertaining to the lowest range of temperatures show a rather well defined initial stage in which the growth rate is almost nil.

By contrast, the samples strained 10% do not show a similar initial stage and, even at the lower annealing temperatures, their mean grain size reaches high values in a short time. On the other hand, for very low rates of strain (~2%), no appreciable increase of linear dimensions is observable (Fig. 7a) up to the highest temperatures, at which the trend of the curves (Fig. 7b) approaches that of the samples strained 5%. Lastly, in the specimens containing inclusions (Armco iron, A), the stage of growth inhibition is extended up to a higher range of temperatures.

While the observations made on the effects of impurities in the preceding paragraph seem not to need further details, a first approach to the discussion of the results on grain-growth kinetics in pure iron may be made by considering Fig. 7a.

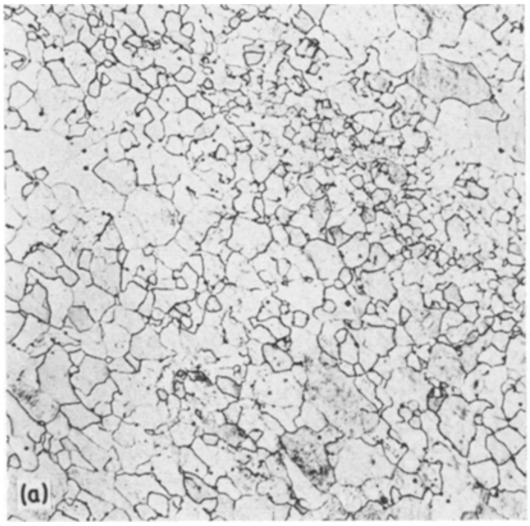
The nearly absolute inhibition of grain-size increase observed for the very slightly deformed specimens (2%) is most probably due, as already remarked, to polygonization, particularly stable at these amounts of deformation.

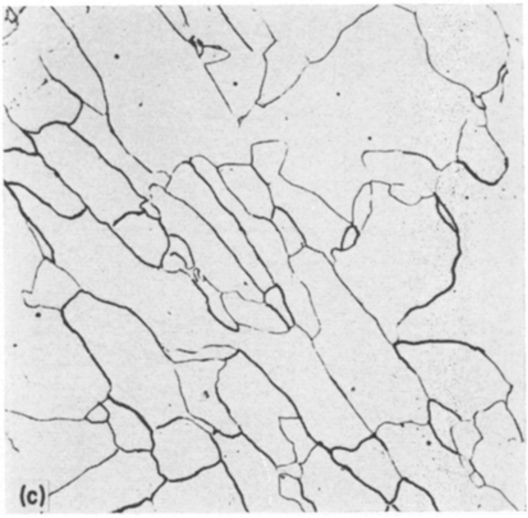
The presence of a polygonized substructure is probably also responsible for the initial stage of growth-inhibition observed for the specimens strained about 5%. At this stage, the lattice of polygonized sub-boundaries acts as a restraining factor against the driving force for grain growth, slowing down the mobility of the primary grain boundaries; in these conditions, a certain period (incubation period) is required for the development of particularly favoured grains which later can behave as nuclei for a secondary recrystallization process, and grow at the expense of the other, smaller grains. At this second stage, a rapid increase of the mean size of grains occurs, giving rise to a sharp knee in the grain size plot versus annealing time. Later on, the rate of growth progressively decreases. Two stages of this process are evidenced in micrographs a and b (Fig. 8) showing the typical pattern of a discontinuous grain growth.

The specimens strained about 10% present an acceleration of the beginning of boundary motion, showing, even from the shortest annealing times at the lowest temperatures, an increase of mean grain size up to very high values. During the successive anneals grain dimensions remain virtually constant or increase slightly. No typical feature of secondary recrystallization can be observed in the grain size distribution, even after the shortest annealing periods (see, for example, Fig. 8c).

The acceleration observed could be attributed partly to the reduction of the restraining effect of polygonization (which is less important in this range of strain than for lower deformations) and partly to the presence of additional driving force, due to the stored energy introduced by deforming the specimens in this range of strain. In the present case (specimens strained 10%), deformation does not produce a strongly deformed matrix; nevertheless local differences in dislocation density from region to region produce a small number of primary nuclei which lead, after annealing, to a large-grain structure [16]. In fact, the appearance of a stage of primary nucleation (not so pronounced however as for higher deformations leading exclusively to primary recrystallization) is observed by means of X-ray techniques (Debye back-reflection).

A kind of competition between primary and secondary recrystallization, and a combination of the two processes, are therefore responsible for the characteristics of grain growth in specimens with low and medium amount of strain. In this range of deformations, for relatively high





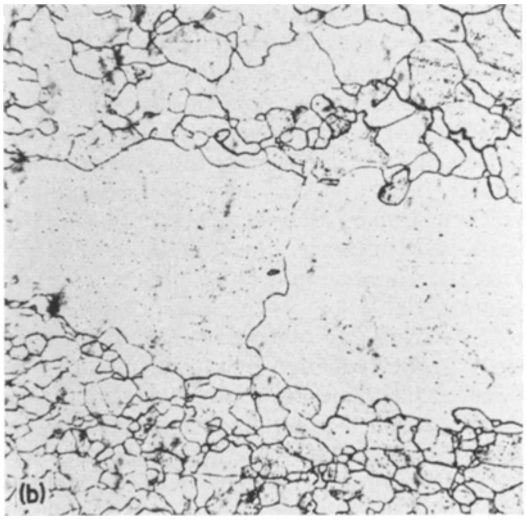


Figure 8 Pure iron (P) strained 5%, annealed at 700° C for (a) 260 min and (b) 800 min. Pure iron (P) strained 10%, annealed at 685°C for 20 min (c). Magnification: (a) ×75; (b) ×65; (c) ×35.

strains (10% and more) primary recrystallization seems to prevail. Only with samples strained about 5%, in our conditions, is it possible to observe the main features of abnormal grain growth both from the standpoint of grain size distribution and of growth kinetics: an initial incubation period with nearly null variation of grain dimensions, followed by a sudden increase at high rates leading to final large grain sizes, is in fact observed.

6. Critical deformation for secondary recrystallization

In Fig. 6a one can observe that the specimen strained 2%, annealed at 776°C, shows grain growth only after a 4 h anneal, while the sample strained 2.4%, though annealed at a lower temperature (755°C), shows a sharp grain size increase even after very short annealing times.

For slightly cold-worked specimens (in our case, about 2%) it seems therefore that very small variations of the amount of strain can strongly affect the aptitude to growth.

In order to define more precisely the exact value of the critical strain limit, some further results, obtained on high purity iron samples (P), strained in tension from 1.74 to 2.40%, are reported in Fig. 9.

It is possible to note that the specimen strained 2.22 and 2.40% already have a sharp grain-size increase at the lowest times of annealing, while the less deformed ones show a very small variation of grain dimensions. After long annealing periods a certain grain growth may be observed even in the latter specimens, but it is much less than in the former two.

An interesting behaviour is provided by the sample strained 2.04%, which is intermediate between the specimen deformed 1.74% and the other more deformed. As shown in Fig. 10, the growth process, when it occurs, consists in a typical secondary recrystallization.

The results here outlined show that a critical strain limit exists, under which polygonization almost completely blocks any boundary motion,

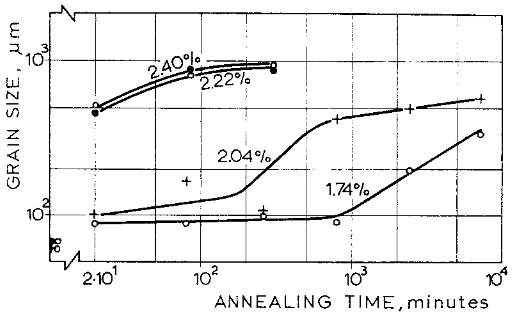


Figure 9 Isothermal grain growth at 755° C for specimens strained in tension. Pure iron (P). The amounts of deformation are indicated.

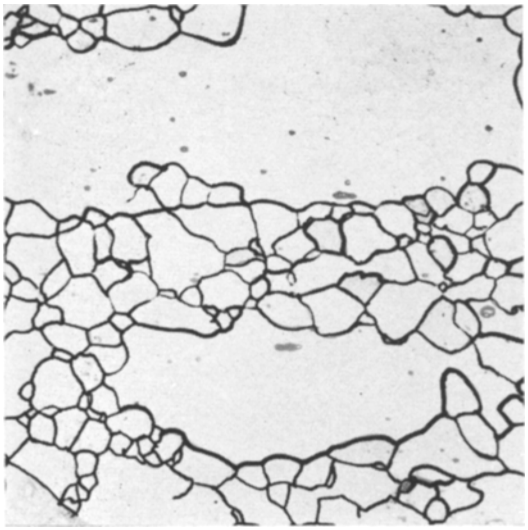


Figure 10 Pure iron (P) strained 2.04%, annealed at 755°C for 260 min (×65).

while above it discontinuous grain growth occurs. In our conditions the critical strain limit was found to be 2.0%; this value must be considered correct at 755°C, but different values are likely at different temperatures, the processes involved being thermally activated.

Other parameters too may influence the value of the critical strain. For instance, as concerns the grain dimensions prior to deformation, interesting observations were made by Morgand [17] on samples of zone-refined iron strained in tension in a range of deformations analogous to that studied in the present work. He finds that specimens with an initial mean grain size of about 10^{-2} mm^2 recrystallize only if deformed in tension by about 8 to 10%, and that below this strain limit polygonization completely blocks any boundary motion. In specimens with a grain size of about 1 mm^2 even for deformations near to breaking point by tension, polygonization remains stable and inhibits recrystallization.

Since the initial grain size of our samples was about 10^{-3} mm^2 and the critical strain by tension about 2%, the hypothesis of a close dependence of the critical limit on the grain dimensions seems acceptable: the larger the initial grain size, the higher the critical limit. Results obtained by other authors [18–22] agree with this hypothesis.

Impurities modify the behaviour of metals deformed at about the critical value, because their presence may favour polygonization, thus inhibiting later recrystallization. Particularly effective is the presence of C, N and O [18, 23], impurities which in our case are practically absent.

7. Conclusions

1. Specimens of high-purity iron after primary recrystallization undergo, when annealed, normal grain growth.
2. Both low amounts of deformation and randomly distributed precipitated oxides retard or inhibit the beginning of grain growth in high-purity iron after primary recrystallization, whilst substitutional and interstitial impurities do not essentially modify the kinetics of the process.
3. Very low elongations (no more than 2% in our conditions) almost completely block boundary motion, by introducing a polygonized substructure.
4. Slightly higher deformations (about 5%) show the characteristic trend of abnormal grain growth; from a kinetic standpoint an initial incubation period with nearly null variation of mean grain size is observed, followed by a sudden, rapid increase of grain size up to the highest values. Only in this case are the typical features of secondary recrystallization evidenced.
5. In a range of relatively higher deformations (10 to 20%), a kind of competition between secondary and primary recrystallization and a combination of the two processes takes place; with respect to the specimens deformed 5%, an acceleration of the beginning of grain growth is observed, but the final mean grain sizes are not very large.
6. In our conditions, 2.0% elongation (at 755°C) is the value of the critical deformation for secondary recrystallization; below this limit

any variation of grain size is practically blocked by polygonization.

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