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New Concepts of Annealing-Twin Formation in Face-Centred Cubic Metals

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The formation and annihilation of annealing twins in oxygen-free copper and in an aluminium bronze has been observed directly and continuously in a photoemission-electron microscope. Taking into consideration both the coherent and the incoherent twin boundaries, it was found that the formation of annealing twins is essentially a typical feature of the recrystallization mechanism whereas the grain-growth process is characterized by the annihilation of these twins, the formation of new twins being comparatively infrequent in this range of annealing. The Fullman–Fisher relationship is not considered to be a valid criterion for annealing-twin formation during recrystallization, and it appears unlikely that the mechanism for twin formation is merely a matter of growth accident, as is widely held.

The model generally accepted for twin formation is that advanced by Fullman and Fisher,[1] according to which a twin can be formed when certain interfacial-energy relationships are satisfied. In addition, concepts have been advanced[2–4] that pertain to the atomic transfer involved in annealing-twin formation and that interpret this formation in terms of growth accidents.

These theories have been based, in most cases, on observations made on materials in which twinning had already taken place, i.e. after the fact. Thus, the process of twin formation, properly speaking, had either not been observed directly or had been studied under poorly controlled conditions. Consequently, a twinned region could rarely be related unambiguously to the same region before twinning, so that a large number of counts had to be taken to obtain a statistical record of the overall behaviour of a given material.

Another serious deficiency of the concepts advanced is the lack of a clear distinction between the range of recrystallization and the range of grain growth. For instance, Fullman and Fisher[1] studied the formation of annealing twins during grain-coarsening only, whereas subsequent workers refer to the Fullman–Fisher model as if it had been proposed to cover both the recrystallization and the grain-growth domain of the annealing sequence. It is, thus, not surprising that results on annealing-twin density are often contradictory; notably, as far as the effects of grain size[5,6] and the amount of cold work[7,8] are concerned.

It is the purpose of this paper to advance new ideas on the dynamics of annealing-twin formation in the light of our observations in the photoemission electron microscope (PEEM).*

*Metioscope KE3 made by Balzers.

Manuscript received 27 September 1972; in revised form 7 February 1973. G. Gindraux, BSc, is Chef des Travaux and W. Form, BSc, FIM, is Professor and Director of the Institute of Structural Metallurgy, University of Neuchâtel, Switzerland.

Scope of Study

Preliminary heating runs in the PEEM indicated that annealing-twin formation during recrystallization differs from that occurring during grain growth. Consequently, a basic study of the dynamics of annealing-twin formation requires that one be able to distinguish between:

twins present in the deformed state;
twins forming during recrystallization;
twins disappearing during recrystallization;
twins forming during grain growth;
twins disappearing during grain growth.

continuously throughout the entire annealing process under consideration.

Furthermore, it appeared to us necessary to take into account not only the coherent but also the incoherent twin boundaries, since it is by the thermally activated movement of the latter that twins can grow or annihilate themselves.

Experimental

Materials and Procedure

For the major portion of the work we used an oxygen-free copper[9] (99·92% Cu) having a stacking-fault energy (SFE) of $\sim$ 70 mJ/m^2 (erg/cm^2) and an aluminium bronze[9] (93·70% Cu, 6·22% Al) with an SFE of $\sim$ 10 mJ/m^2 (erg/cm^2). In each case the material was received in the form of extruded bars, 15 mm in dia.

These materials were subjected to a homogenizing treatment of 770° C for 15 h in a salt bath and then deformed by 30% reduction in area in a swager. From this 'initial' state onwards, the thermal and mechanical treatments differed between the various samples analysed, depending upon the grain size and amount of cold work introduced before the heating run in the PEEM.

Specimen Preparation and Heat-Treatment in the PEEM

The operation of the Metioscope KE3 has been described in various papers,[10,11] and also the method of specimen preparation.[12,13]

All thermal treatments in the PEEM were carried out in the following sequence:

(i) rapid heating (at ~ 50 degC/min) to ~ 10 degC below the temperature of the beginning of recrystallization, as determined in a dry run;

(ii) passing through the recrystallization range at ~ 1 degC/min. (It must be emphasized that we did not attain structural equilibrium of the recrystallizing matrix in this way[14] but only thermal equilibrium): at least 10 pictures were taken in this range;

(iii) heating into the grain-growth domain in steps of 50 degC, the specimen being held at each step for 30 min before a picture was taken.

Thanks to an important modification applied to the Metioscope[14] it was possible to maintain the temperature at ± 2 degC, the degree of precision necessary to perform the type of study in question.

Counting Technique

The same area of a specimen surface is studied during any one heating run in the PEEM. Consequently, the number of counts is somewhat limited. Observations were made on 50 × 90 mm plates at a magnification of 440 ×. Thus, the number of events that can be followed on the fluorescent screen of the PEEM is restricted. The types of twin observed have been classified in the manner illustrated in Fig. 1.

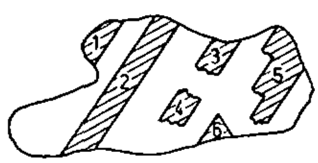

Fig. 1 Types of twin observed:
(1) 1 *coherent boundary*
(2) 2 *coherent boundaries*
(3) 2 *coherent boundaries and* 1 *incoherent boundary*
(4) 2 *coherent and* 2 *incoherent boundaries*
(5) 2 *coherent boundaries and* 1 *incoherent boundary, one of the coherent boundaries being continuous and the other interrupted by steps of the incoherent boundary*
(6) 2 *coherent boundaries*

We did not determine the 'twin density' but rather counted the number of coherent and incoherent twin boundaries in a given area of the picture taken in the PEEM.

The pictures taken in the recrystallization range were subsequently magnified to 1320 ×. The recrystallized area was then determined with a planimeter on each micrograph. Finally, the number of coherent and incoherent twin boundaries within this area was counted and related to the percentage of recrystallized area.

One important aspect must be emphasized here. Earlier researchers were not always able to distinguish unambiguously between the twins in the deformed and those in the recrystallized area in the temperature range and over the time range where recrystallization took place. Consequently, their reported 'twin densities' include both the twins already present in the deformed matrix and those formed in the course of recrystallization. This mixing of old and new twins[8] provides, of course, a confusing picture of actual twin formation. Our counts, on the other hand, include only those twins that have been newly formed in the recrystallized region, thus affording a clear picture of twin formation during annealing.

In the temperature range of grain growth the procedure was somewhat different. Since one cannot readily quantify 'grain-coarsened' areas, we counted the change in number of coherent and incoherent boundaries, starting from the fully recrystallized state, and related the results to grain-coarsening temperature.

We have estimated the error inherent in our counting method to be ~ ± 10%.

Results

The results presented relate to heating runs made on Cu-OF in four different metallurgical states, as characterized by grain size and cold work, and on one 6% aluminium bronze. The necessary thermal and mechanical treatments to achieve these states were applied to the material in the 'initial' state as specified above. Table I lists these metallurgical states.

TABLE I
Grain Size and Amount of Cold Work (by Swaging) before Heat-Treatment in the PEEM

Material	Designation	Av. Grain Size, μm	Amount of Cold Work, %
Cu-OF	CA 41	36	11·0
	CA 42	80	26·4
	CA 43	36	26·4
	CA 44	80	11·0
Cu-6% Al	—	100	24·0

It should be emphasized that our results relate exclusively to surface observation and, furthermore, that no attempt was made to convert them into volumetric terms on the basis of an assumed simple grain shape.

From the total of roughly 120 pictures we include here a representative set of 8 micrographs taken in the course of the heating run of material CA 43. Fig. 2 shows the variation in temperature with time for this series and enables the electron micrographs to be correlated with the appropriate annealing range.

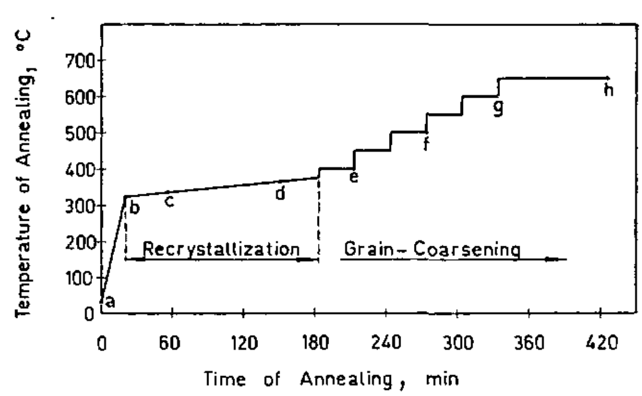

Fig. 2 Time/temperature relations during the heating run of sample CA 43, The small letters refer to the pictures in Fig. 3.

(a) (e)

(b) (f)

(c) (g)

(d) (h)

Fig. 3 Electron micrographs of sample CA 43 taken in the PEEM during recrystallization and grain-coarsening. The time and temperature at which these pictures were taken can be obtained from Fig. 2. (The recrystallized areas are encircled by a black line in Figs. 3(b) *and* (c)).

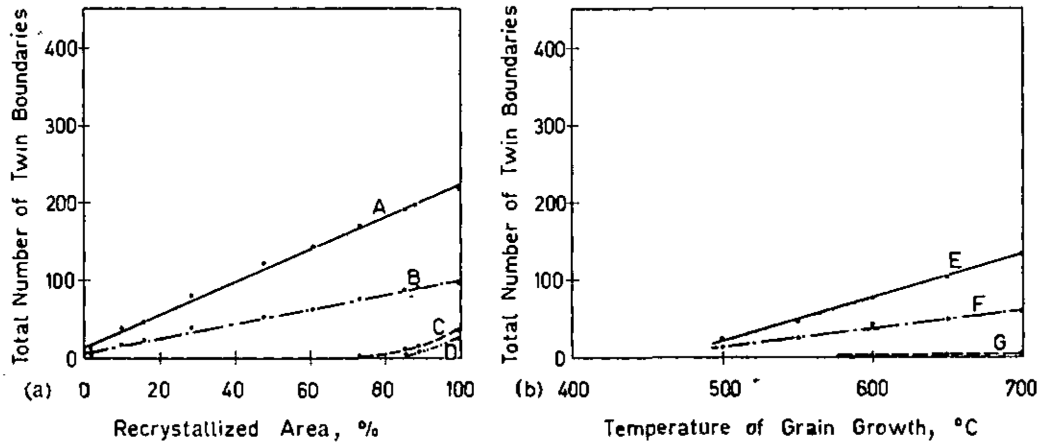

Fig. 4 *Formation and destruction of twin boundaries in CA* 41:
(a) *during recrystallization;* (b) *during grain growth.*
A and G: formation of coherent twin boundaries
B: formation of incoherent twin boundaries
C and E: annihilation of coherent twin boundaries
D and F: annihilation of incoherent twin boundaries

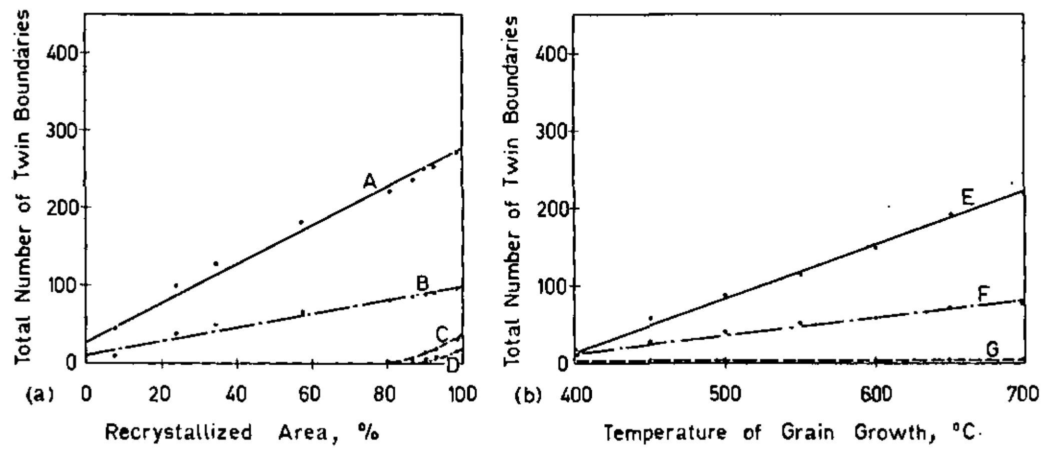

Fig. 5 *Formation and annihilation of twin boundaries in CA* 42:
(a) *during recrystallization;* (b) *during grain growth. Key as Fig.* 4.

Fig. 3 contains the selected eight pictures of the CA 43 run. In the as-deformed state (*o*) the orientation contrast is rather difficult to obtain, giving the micrograph a somewhat hazy appearance. In the grain-coarsening range, on the other hand, thermal etching clearly outlines the previous positions of grain boundaries. The recrystallization range is illustrated by (*b*)–(*d*), whereas (*e*)–(*h*) represent the grain-coarsening range. The results obtained from the counts of the coherent and incoherent twin boundaries in the regions of recrystallization and of grain growth are reproduced in Figs. 4–8. The straight lines in these diagrams are regression curves obtained from the best least-squares fit, as calculated by computer.

It should be noted that the area analysed for the aluminium bronze was 3·9 times smaller than that for the copper runs, so that the values for the former material must be multiplied by this factor if a comparison between the two types of material is to be made.

For the counts in the recrystallization range the deviation to the ordinate from the origin of the coordinate system reflects the error inherent in our counting technique.

From these results one may draw the following main conclusions:

(1) The formation of annealing twins occurs mainly during recrystallization, whereas it is a rather infrequent event during grain-coarsening.

(2) The destruction of newly formed annealing twins is negligible during recrystallization, but is the dominant process during grain growth.

(3) The proportion of incoherent relative to coherent twin boundaries is much higher during recrystallization than in the grain-coarsening region, i.e. twins that traverse a new grain only partially are typical of the recrystallized state.

Furthermore, from the straight-line relationship in Figs. 4–8 one may conclude that:

(4) A given increase in amount of recrystallization produces the same number of new coherent and incoherent interfaces, irrespective of the recrystallization interval and of the particular surface region within which recrystallization takes place.

(5) The number of twin boundaries annihilated during grain growth in a given temperature range is independent both of the temperature interval in which annihilation takes place and of the particular surface region in which grain-coarsening proceeds.

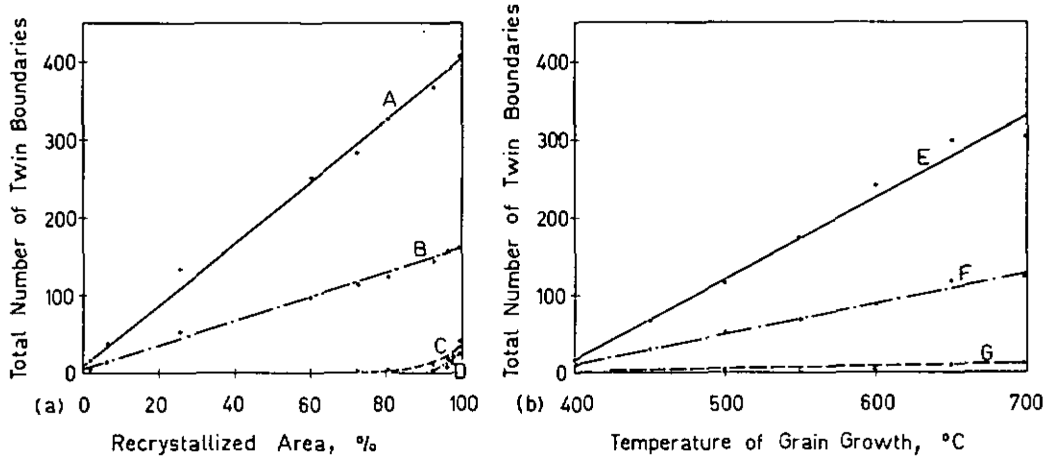

Fig. 6 Formation and annihilation of twin boundaries in CA 43: (a) during recrystallization; (b) during grain growth. Key as Fig. 4.

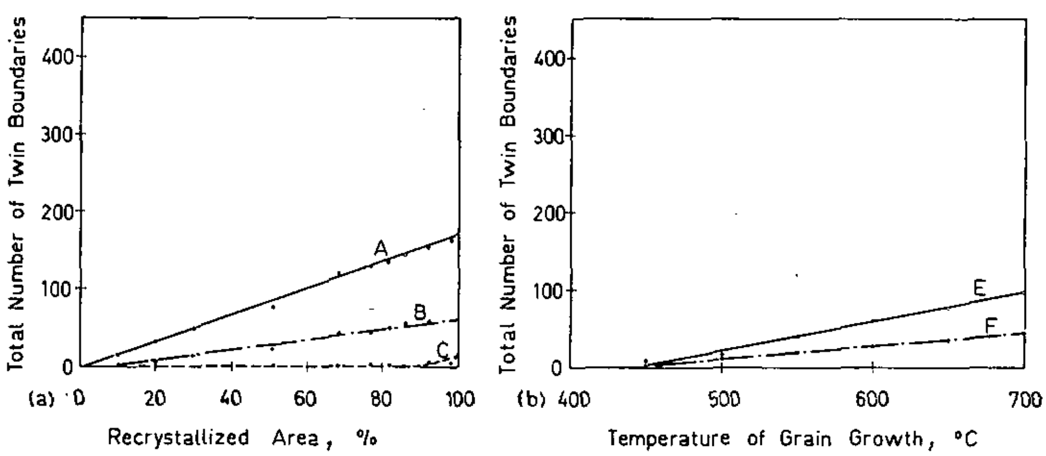

Fig. 7 Formation and annihilation of twin boundaries in CA 44: (a) during recrystallization; (b) during grain growth. Key as Fig. 4.

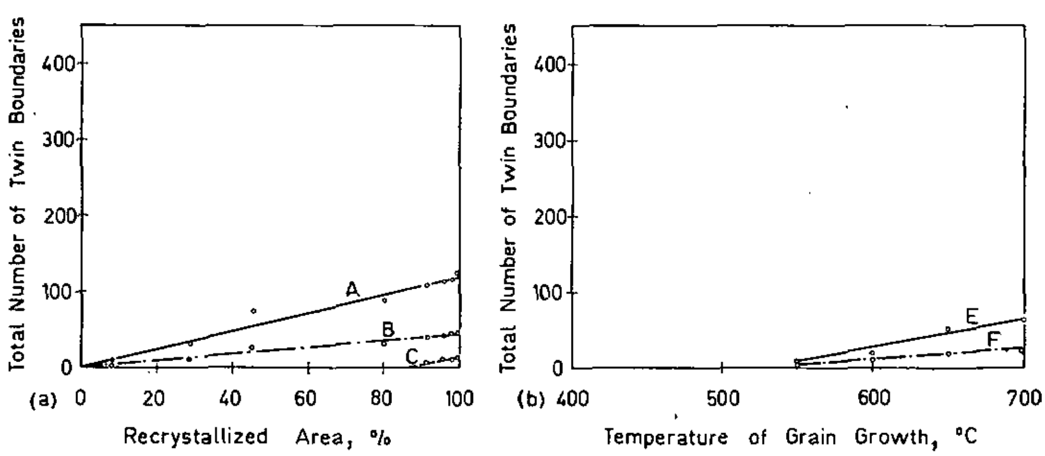

Fig. 8 Formation and annihilation of twin boundaries in Cu–6% Al: (a) during recrystallization; (b) during grain growth. Key as Fig. 4.

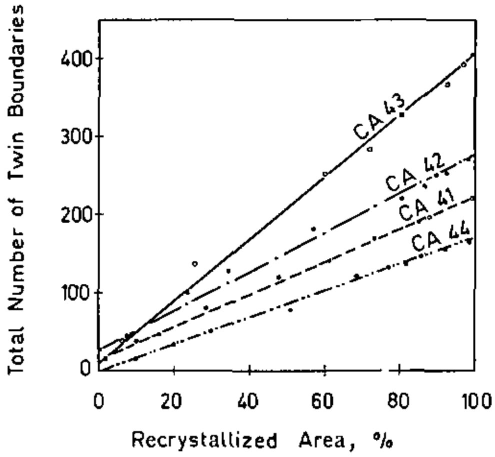

Fig. 9 Formation of coherent twin boundaries during recrystallization in the four copper samples analysed.

Also, when comparing the four heating runs on copper with one another (Fig. 9), one notes that the number of coherent twin boundaries is greatest in the material with the smallest average grain diameter in the deformed state and the highest amount of cold work (CA 43), whereas it is lowest in the material with the largest initial grain and the least reduction in area (CA 44), the other two samples being characterized by intermediate numbers of twin boundaries.

Finally, correcting the values for the aluminium bronze to bring them on to the same basis of comparison as CA 42 (the copper sample most closely resembling it in grain size and amount of cold work), one tends to conclude that lowering the SFE does indeed increase the number of coherent twin boundaries, as is widely held.

Discussion

Energetic Considerations

Our results clearly support the view that, in the metals studied, the formation of annealing twins is above all a phenomenon accompanying the recrystallization process, whereas during grain-coarsening most of the twins created during recrystallization are annihilated and the formation of new twins is the exception rather than the rule. These observations are in disagreement with results reported by other researchers[1,4,8,15] and none of the models so far advanced can account for twin formation during recrystallization.

Let us recall that the driving force for recrystallization is the energy stored in the cold-worked state, mostly in the form of dislocation energy. Recrystallization thus constitutes that phase of annealing during which the dislocation density is reduced to an extremely small fraction of the amount present in the initial, cold-worked material by the movement of newly formed grain boundaries.[14] Yet, in quantitative studies of twin formation conducted on the same materials as those used here[15] this driving force is not included.[4,8,15] Moreover, the model of Fullman and Fisher,[1] which describes only an equilibrium condition, also does not apply to twin formation during recrystallization since the recrystallized matrix is not in an equilibrium state. Rather, it represents an intermediate stage between the cold-worked state and that which may ultimately be reached by grain-coarsening.

On the other hand it is known that the reduction of the elastic energy stored in the cold-worked material must occur along that reaction path by which the free energy can be reduced most rapidly. In line with this elementary principle, we propose that the role of annealing twins is analogous to that played by the deformation twins: they reorient the recrystallizing matrix in order to permit a continued rapid reduction in the stored energy, in the same way as the deformation twins reorient a portion of a crystal in order to facilitate continued plastic flow by slip.

Looking at the energetic balance for recrystallization one can write for the total driving force, E_{rec}, in a given sample:

$$\Delta E_{rec} = \Delta E_{el} + (\Delta E_{gb})_o + (\Delta E_{tb})_o - (\Delta E_{gb})_n - (\Delta E_{tb})_n$$

where

ΔE_{el} = stored elastic energy
ΔE_{gb} = grain-boundary energy
ΔE_{tb} = twin-boundary energy

o, n refer to the old and the newly formed boundaries, respectively.

During recrystallization of our samples all the old grain and twin boundaries were completely eliminated. The dominant term on the right-hand side in the above energy relation is ΔE_{el}, which is precisely the term neglected in considerations of twin formation made hitherto. In fact, this term is so much larger than any of the other terms that one could write:

$$\Delta E_{rec} \simeq \Delta E_{el}$$

Under such conditions a local increase in energy through the formation of a low-energy annealing twin (< 1% of ΔE_{el} in terms of J/mol) is entirely possible, if continued rapid reduction in stored energy requires a reorientation of the crystal.

As far as the grain-coarsening region is concerned, the driving force, ΔE_{gg}, can be formulated as follows:

$$\Delta E_{gg} = (\Delta E_{gb})_n + (\Delta E_{tb})_n - (\Delta E_{tb})_{n'}$$

where n refers to the recrystallized state, and n' to possible twin formation during grain growth.

The dominant term on the right-hand side of this relationship is $(\Delta E_{gb})_n$, of which $(\Delta E_{tb})_n$, may, nevertheless, constitute an appreciable fraction.[8] Since the driving force ΔE_{gg} is very small (of the order of 1% or less of ΔE_{rec}), there is no obvious reason why a local increase in energy of substantial relative magnitude should facilitate grain growth. Of course, since after recrystallization the dislocation density is still $\sim 10^6$ cm^{-2}, it is possible that through the movement of grain boundaries local dislocation agglomerations can occur, the rapid elimination of which may be enhanced by a local reorientation of the lattice. Such an interpretation would then attribute twin formation during recrystallization and during grain growth to the same basic cause, but the frequency of the occurrence of the latter would naturally be small.

What follows from our observations is that the deformed state (in the sense of high local dislocation densities) is a necessary—but not sufficient—condition for annealing twins to form. To check this point we have examined a number of as-cast materials including an α-brass, a nickel silver, and a free-machining copper. In no instance were any twins observed. After reheating the as-cast copper to 600° C for 2 h we noted some slight grain-coarsening, but no twin formation. Only after cold swaging the as-cast copper 35% and annealing at 600° C for 1 h were some twins present. These results are in contradiction to those of Charnock and Nutting[8] obtained on as-cast material. Possibly, their technique of chill-casting was severe enough to produce plastic flow at a temperature high enough to induce recrystallization.

Another point left unexplained by our results relates to those studies[1,4,8,15] in which the twin density or frequency was found to increase with temperature, the latter being presumably situated within the grain-coarsening range. One distinct possibility of explaining the discrepancy between these results and our own may lie in the choice of different parameters to evaluate the twinning tendency. For instance, when expressing twin density in terms of twins per grain it is possible that during grain-coarsening this density increases, though the total number of twins diminishes and no new twins are formed. To this ambiguity one must add the fact that some researchers did not distinguish between twins formed during an earlier interval and those formed during the subsequent period of heat-treatment. These difficulties and uncertainties have been eliminated by our method of observation and counting.

Effect of Grain Size and Cold Work

It has been found that for a relatively small amount of prior cold work (11%) the difference between an initially fine and an initially coarse grain was small as far as the formation and the annihilation of twin boundaries is concerned (Figs. 4 and 7). For larger amounts of deformation, on the other hand, the grain-size effect is more pronounced (Figs. 5 and 6), in the sense that the number of twin boundaries formed is higher for a finer grain.

The interdependence of grain size and cold work, as far as their influence on the tendency to form annealing twins is concerned, becomes evident from a comparison of the 4 heating series conducted on the Cu-OF (see Fig. 9). This figure suggests that the same tendency to twin formation can be induced in a material through a variety of different combinations of grain size and cold work. As a matter of fact, it appears that the number of coherent twin boundaries is related to the dislocation density, which, in turn, is related to grain size and deformation by the well-known equation

$$\varepsilon = b\,\bar{x}\,\rho$$

where ε = macroscopic shear

$\bar{x}$ = mean free dislocation path (which may be considered here to be proportional to grain size)

ρ = dislocation density

It follows directly from this relationship that the larger the grain size the lower is the dislocation density induced by a given macroscopic deformation and vice versa.

From the results in Fig. 9 one may deduce that the driving force for twin formation varies in the same sense as that for recrystallization, the latter too, being governed primarily by the elastic energy stored in the form of dislocations. In Fig. 10 the electron micrographs of the four copper samples are displayed at the moment when they attain 100% recrystallization. The micrographs are arranged in the order of what is presumed to be an increasing dislocation density. We observe that the recrystallized grain size diminishes, whereas the total number of twins increases. This is precisely the order to be expected on the basis that one of the principal parameters governing twin formation in a given material is the dislocation density present in the cold-worked state, irrespective of the initial grain size and cold work.

For the two samples containing the highest (CA 43) and the lowest (CA 44) dislocation density, respectively, it was found that for the average recrystallized grain size:

$$\text{CA 43 } (d_{rec})_{av} \simeq 27\ \mu\text{m}$$
$$\text{CA 44 } (d_{rec})_{av} \simeq 63\ \mu\text{m}$$

and for the total number of coherent twin boundaries:

$$\text{CA 43} \simeq 400$$
$$\text{CA 44} \simeq 180$$

From these results one may deduce that the ratio of coherent twin boundaries in two different samples is inversely proportional to the ratio of the respective diameters of the recrystallized grains, irrespective of the amount of prior cold work and initial grain size. This is a highly significant result which should be investigated further since, if it proved to be of general validity, it would throw considerable light on the mechanism of annealing-twin formation.

A final word on the role played by the stacking-fault energy (SFE). Although the corrected value for the aluminium bronze (Fig. 8) indicates that this alloy, with the lowest SFE, has the highest total number of coherent twin boundaries in any of the heating runs carried out, one notes that the corresponding value is not far in excess of that for the copper specimen CA 43, which has an SFE seven times that of the aluminium bronze. This clearly shows that the SFE has no direct bearing on the twin frequency, as implied by the Fullman and Fisher relationship,[1] as well as other quantitative treatments.[4] It appears more likely that the SFE participates only indirectly in annealing-twin formation in that it controls the stored energy, ΔE_{el}, and the arrangement (planar, cellular, polygonal, random) of the dislocations in the cold-worked state which, in turn, governs the twin formation in the sense described above.

Conclusions

Thanks to the PEEM we have been able to clarify several basic aspects of annealing-twin formation, without specifically referring to its atomistic mechanism. For the case of oxygen-free copper and a 6% aluminium bronze it has been shown that:

(1) The formation of annealing twins occurs to an overwhelming extent during recrystallization of a deformed matrix. The probability that it proceeds in a non-deformed matrix appears extremely slight. This has been demonstrated for as-cast materials, as well as for the grain-coarsening stage.

(2) The formation of twins requires the movement of newly formed grain boundaries. In no case was it observed that the boundaries already existing in the deformed matrix started to move to permit twin formation.

(3) During recrystallization a small proportion of the newly formed twins disappear again.

(4) During grain growth the twins are progressively annihilated, either through the sweeping over of a grain boundary, or through 'self-destruction' by movement of the non-coherent twin boundary. The extent of formation of new twins during this annealing phase is very small.

(5) The effects of initial grain size and prior cold work on twin formation are related in the sense that these two parameters govern the dislocation density, which appears to be

(*a*) (*b*)

(*c*) (*d*)

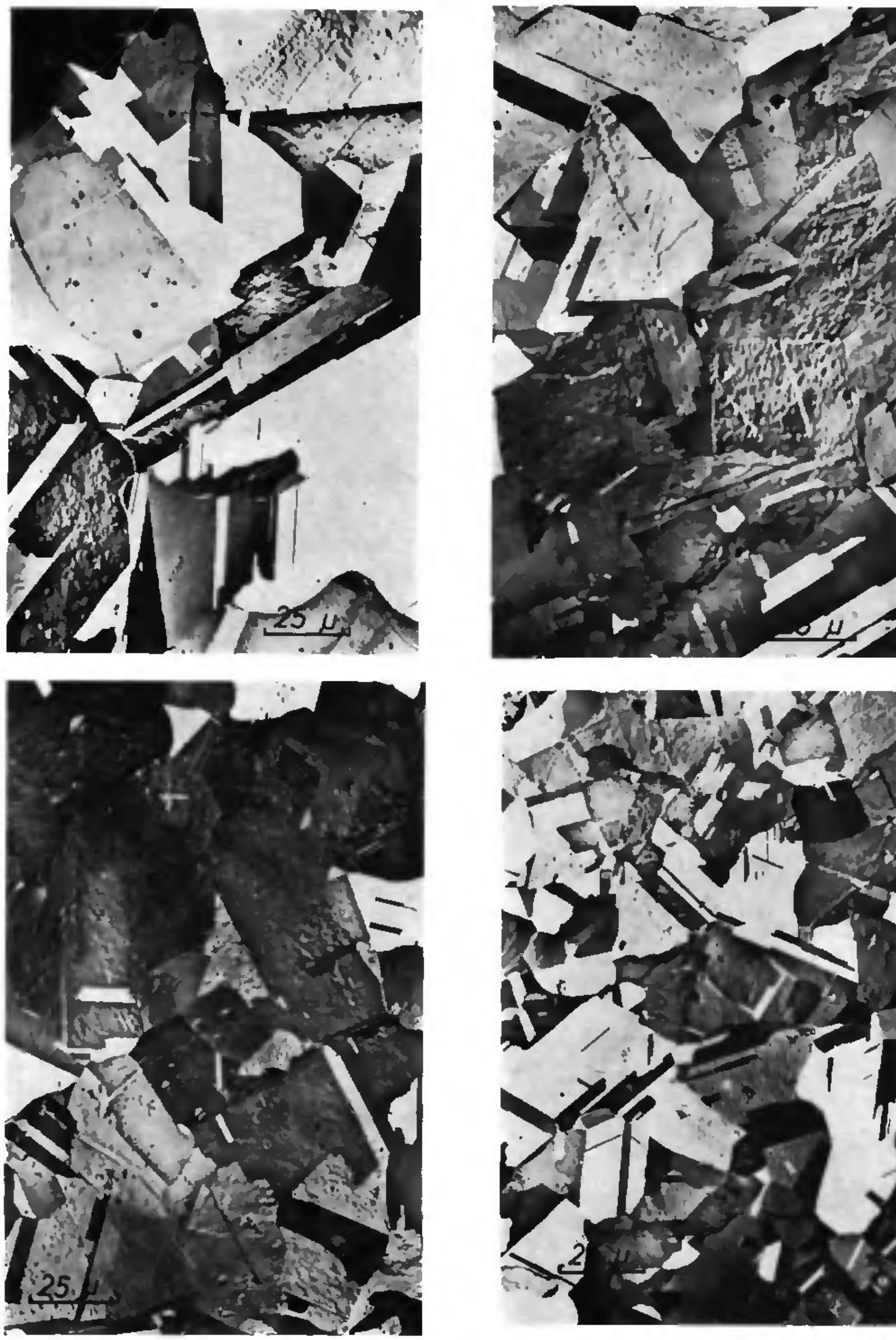

Fig. 10 *Electron micrographs of the four copper samples in the fully recrystallized state. The pictures are arranged in the order of decreasing grain size:* (a) *CA* 41; (b) *CA* 44; (c) *CA* 42; (d) *CA* 43.

one of the controlling factors for annealing-twin formation in a given alloy.

(6) A marked variation in SFE leads only to a relatively small variation in twin density. At any rate, on reducing the initial grain size by a factor of ~ 3 (CA 43), one obtains only a slightly lesser number of coherent twin boundaries than by reducing the SFE by a factor of 7. This strongly suggests that the role played by the SFE consists, above all, in the determination of the density and arrangement of the dislocations before the annealing treatment, and not in the satisfaction of any type of surface-energy relationship.

(7) It appears that, independent of initial grain size and amount of cold work, the ratio of the number of twins in two samples is inversely proportional to the ratio of the corresponding recrystallized grain sizes.

Our results clearly refute the theory of Fullman and Fisher—not merely for the simple reason that their model is based on an equilibrium condition never attained during recrystallization. We propose that the formation of annealing twins—in analogy with the formation of deformation twins—is a means of reorienting part of the structure so as to permit, during annealing, the most rapid reduction of that energy which is stored in the deformed matrix mostly in the form of dislocations.

Acknowledgements

The authors gratefully acknowledge the financial support of the Swiss Foundation for Scientific Research, which permitted the acquisition of the Metioscope KE3. Thanks are also due to the Association Métallurgique SA, Berne, which provided a fellowship for one of them (G.G.).

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Cette publication résume la thèse de doctorat soutenue à l'Université de Neuchâtel par Gilbert Gindraux le 14 décembre 1972.