

# New Concepts of the Recrystallization Phenomenon

M. Schweizer and W. Form

The course of recrystallization in a pure copper and in an aluminium bronze has been observed directly and continuously in a photoemission electron microscope. It could be shown that a range of temperatures exists—termed the 'range of partial recrystallization'—within which recrystallization does not go to completion when carried out at constant temperature. Consequently, the generally accepted idea that an Avrami-type equation best describes the kinetics of recrystallization must be considered to be incorrect. It is also observed that it becomes more difficult for recrystallization to attain completion as prior deformation becomes greater, which is contrary to general belief. As far as average grain size is concerned, the widely held view, that under ordinary heating rates this increases with recrystallization temperature, has not been confirmed, as no such variation was observed. It is indeed meaningless to talk about average grain size within the partial recrystallization range, since here the deformed grains of the old matrix coexist with recrystallizing grains. Finally, bulging out of an existing grain boundary to initiate recrystallization, as proposed in the theory of Bailey and Hirsch (*Proc. Roy. Soc.*, 1962, [A], 267, 11), was not observed in any of the cases studied, and seems an unlikely model for the nucleation mechanism of recrystallization.

The concept of recrystallization as a recovery process in cold-worked materials is implicit in the great majority of the heat-treatments carried out in many important industrial manufacturing processes. The first study of its nature goes back about one hundred years,<sup>1</sup> whereas a quantitative assessment of the kinetics has only been undertaken since 1939.<sup>2,3</sup>

In view of its long tradition and practical significance one might assume that the phenomenon of recrystallization is well understood. Indeed, publications in this field are numerous<sup>4</sup> and suggestions as to what is supposed to happen during the corresponding reorganization of the atomic structure are plentiful. Yet, when analysing critically the pertinent literature one comes to the conclusion that this is not the case. Specifically, arguments advanced in support of nucleation theories<sup>5,6</sup> and recrystallization kinetics<sup>7-9</sup> are, to say the least, not very convincing if not actually incompatible with experience.

One of the main reasons for this state of affairs may be found in the fact that direct and continuous observation of the structural changes occurring during recrystallization has not been possible up to now. Therefore, the assessment of recrystallization kinetics has had to be made by means of 'static' observation, i.e. before and after the event, and this not only on one but on a series of specimens, which usually did not bear a clear-cut relationship to one another.

The photoemission electron microscope (PEEM), which has recently become available,<sup>10</sup> permits the desired direct and continuous observation of structural changes in optically isotropic specimens, and this at temperatures that lie well below the thermionic emission temperature of the metal or alloy to be examined. Such an instrument is particularly valuable for the study of 'civil-type' transformations, where no shape change occurs.<sup>11,12</sup> The most important aspect of PEEM is the orientation contrast produced by the photoemission of the electrons, so that the details of the grain structure are revealed without the specimen being chemically

etched.<sup>11</sup> This is the indispensable requirement for the continuous observation of structural changes in optically isotropic materials occurring by the movement of an incoherent interface.

The PEEM is thus the appropriate tool for the direct study of the recrystallization kinetics of specific localized areas of a metallic sample. It permits, above all, the establishment of quantitative phenomenological relationships without necessarily offering any direct information as to the details of the underlying mechanism or the nucleation process. For this latter aim to be achieved the PEEM must be employed in conjunction with the transmission electron microscope (TEM). This will be done in a future phase of the work now presented. The results reported here are of a phenomenological character only, yet they are sufficiently significant to cast serious doubt on generally accepted theories of recrystallization kinetics.

## Background of the Study

It is widely held<sup>5,7,13-16</sup> that the kinetics of recrystallization can be described by an equation of the type<sup>2,3</sup>

$$\eta = 1 - \exp(-Bt^n)$$

where  $\eta$  = fraction of the recrystallized volume  
 $B$  = a temperature-dependent factor  
 $t$  = time at temperature  
 $n$  = a constant

This equation implies that recrystallization is an isothermal process in the sense that within the temperature range in which it can occur, it should go to completion at any temperature, if a sample is held there long enough.

Yet preliminary heating experiments carried out in the PEEM strongly suggested that this might not be the case. Also, there is at least one indication in the literature<sup>17</sup> that recrystallization in a specimen may proceed only partially at a given temperature, despite the fact that the sample in question is held for a very long time at that temperature.

Manuscript received 13 July 1972; in revised form 10 October 1972. W. Form, DSc, FIM, is Professor and Director of the Institute of Structural Metallurgy, University of Neuchâtel, Switzerland, where the work was carried out. M. Schweizer, DSc, is now with the Research Group, Boillat Brass Corp., Neuchâtel.

TABLE I  
Heating Runs on Cu-10% Al and Cu (ofhc) in the PEEM

Run No.	Material	Reduction in Area, %	$T_{e1}$ , °C	$t_1$ , h	$T_{e2}$ , °C	$t_2$ , h
1	Cu-Al	7.7	410	5.2	415	5.1
2	Cu-Al	11.3	410	5	415	5
3	Cu-Al	17.1	415	4.8	420	5
4	Cu-Al	17.1	410	5	425	5.2
5	Cu-Al	21.9	415	6	420	4.3
6	Cu-Al	21.9	425	5.1	430	5
7	Cu-Al	21.9	410	5	433	5.1
8	Cu-Al	26	405	5	415	4.8
9	Cu-Al	26	425	5.2	430	4.7
10	Cu (ofhc)	26	347	4.8	357	5
11	Cu (ofhc)	26	326	4.9	340	5.1

However, it would seem that the significance of this result was not recognized by the authors concerned.

One of the aims of our study was to verify whether or not a temperature range of partial recrystallization existed and, if so, to identify it clearly.

### Experimental

#### Materials

It was at first intended to study the recrystallization kinetics of both phases in a two-phase alloy. However, when it was realized that the basis of recrystallization was poorly understood, we decided to begin our work with the analysis of what happens in single-phase alloys. Since by this time we had mastered the polishing technique for a particular two-phase alloy, namely a 10% aluminium bronze, it was decided to keep to this alloy, for the following reasons:

It satisfied the conditions imposed by the PEEM (i.e. it does not contain any elements of high vapour pressure).

The phase boundaries in the two-phase region are practically vertical,<sup>18</sup> so that no change in proportion of the coexisting phases would occur during the intended heat-treatments.

The second 'phase', present in the form of an eutectoid, does not participate in any perceptible way in the recrystallization process in the range in which the  $\alpha$  phase recrystallizes.

Nevertheless, to verify whether the principal results obtained with the Cu-10% Al alloy could also be obtained with a true single-phase alloy, a heating run in the PEEM was also carried out with an oxygen-free, high-conductivity copper.

The composition of the aluminium bronze was as follows: Cu 90.03, Al 9.8, Pb 0.01, Fe 0.04, Mg 0.01, Si 0.01, Ni 0.01, remainder 0.09 wt.-%. This material was received in the form of extruded and drawn bars of 10 mm dia. Cylinders 25 cm long were machined to a diameter such that, upon subsequent deformation in a swaging machine (to a dia. of 7 mm), the desired percentage reduction was achieved. Before swaging, these cylinders were homogenized in a salt bath for 15 h at 725° C, quenched in a second salt bath at 540° C, and stabilized there for 5 h. After this treatment the alloy contained ~ 33% eutectoid.

The bars were then swaged to 7.7, 11.3, 17.1, 21.9, and 26% reduction in area. The radial strain gradient produced in this way was of no consequence as regards the intended purpose, since the area of observation in the PEEM was always at or very close to the centre of the cross-section. The samples for the heating runs in the PEEM were in the form of cylinders 7 mm in dia. and 5 mm in height.

Since, for the reasons given above and explained elsewhere,<sup>11</sup> the specimen surface to be observed could not be etched, the polishing procedure necessary to develop the desired orientation contrast required special techniques and the utmost care. As the depth from which excited electrons can reach the free surface and then be accelerated to the anode under 40 kV is very shallow (< 100 Å) any deformed layer or thin film deposited on the surface must be kept to an absolute minimum.

The specimens were mechanically prepolished and subsequently finished electrolytically using extremely short current pulses under high tension. After introduction into the PEEM the specimens were always cleaned with an ion beam at 90° incidence, whereas during observation, while being heated in the PEEM, the samples were cleaned continuously or sporadically with a tangentially positioned ion gun.

#### Procedure

To locate the proposed temperature band of partial recrystallization, a very close control over the specimen surface temperature and the ability to keep the latter constant within narrow limits, are indispensable requirements. This condition was not fulfilled with the PEEM, necessitating modifications; these were carried out and finally enabled the temperature to be kept to  $\pm 2$  deg C. This is an unusually high precision in temperature control, never reached in a TEM, for instance. It should be emphasized that, without the realization of this precision, the primary aim of our study could never have been achieved.

It may also be mentioned in this context that heating was accomplished by means of a resistance coil surrounding the specimen-holder and permitting a temperature of up to 1000° C to be reached. Through dry runs the approximate temperature at which recrystallization could first be detected, and at which the first isothermal observation was to be made in subsequent long-time runs, was determined.

Each specimen was analysed at two temperatures. They were all heated first at a constant rapid rate to ~ 10 deg C below the temperature,  $T_{e1}$ , of the first isothermal run and then raised to this latter temperature in a few minutes. Observations at  $T_{e1}$  were made until—watching the fluorescent screen—no increase in recrystallized area could be discerned optically within a period of not less than half an hour. During this period pictures were taken at intervals of 30 min on plates measuring 6 × 9 cm. A magnification of ×345 in

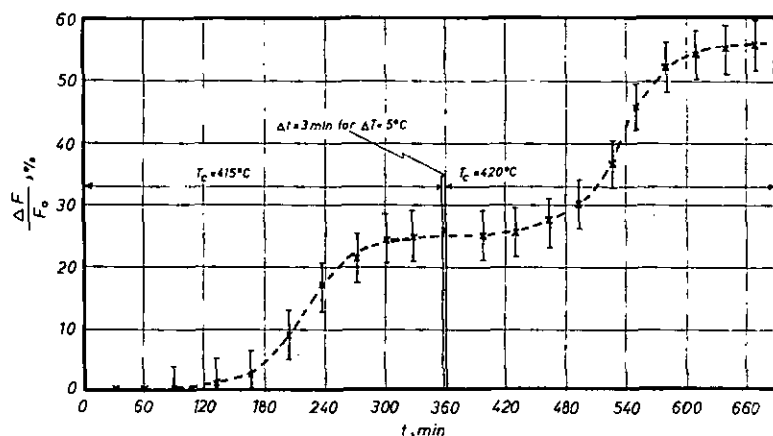


Fig. 1 Change in recrystallized area as a function of time at two temperatures (21.9% prior deformation).

the PEEM was chosen, such that  $\approx 15$ –20 grains were visible on each plate.

The temperature was then raised in a few minutes to  $T_{c2}$  and the procedure described above repeated. The temperature runs are detailed in Table 1.

The times  $t_1$  and  $t_2$  indicate the length of the period during which observations at  $T_{c1}$  and  $T_{c2}$ , respectively, were made.

The quantitative analysis of the plates was carried out as follows: the plates were first copied at an additional magnification of  $\times 3$  yielding micrographs of a size that permitted an area determination of sufficient accuracy. On these photographs the area of the unrecrystallized  $\alpha$  phase was first measured. Subsequently, the recrystallized portion of this phase was measured<sup>11</sup> at a given time, fixed by the instant of taking the picture at  $T_c$ . For this purpose a coordinate net was used. The area to be determined was approximated as closely as possible by a polygon and then calculated from the coordinates of its corner points employing a small computer. Using a line width of 0.1 mm, the calculated area was obtained with an accuracy of  $\pm 2\%$ . If this value is added to the scatter due to the choice of the particular polygon, a total error is involved for the determination of the recrystallized area of  $\sim \pm 3\%$ .

## Results

### The Saturation Phenomenon

Fig. 1 shows a typical series of evaluations of the recrystallized area as a function of time at  $T_{c1}$  and  $T_{c2}$  after a reduction in area of 21.9%. It may be noted that at  $T_{c1} = 415^\circ\text{C}$ , the change in recrystallized area after 294 min is  $< 1\%$  over a period of half an hour. This clearly proves that recrystallization tends toward a saturation value, and applies even though  $< 25\%$  of the total  $\alpha$ -phase region had recrystallized. A similar trend is evident at  $T_{c2} = 420^\circ\text{C}$ . Moreover, after raising the sample to this latter temperature, recrystallization does not continue immediately but only after an appreciably long delay time.

The results obtained in heating runs 10 and 11 clearly show that partial recrystallization occurs also in the oxygen-free pure copper. As is to be expected, copper exhibits a somewhat lower range of recrystallization temperature than the Cu–10% Al alloy that has undergone the same prior reduction in area.

Fig. 2 shows portions of the micrographs taken during run 3. The saturation tendency is particularly evident on comparing Fig. 2(j) with Fig. 2(k), since not the slightest change in recrystallized area can be detected between the two.

It should be pointed out that a somewhat different evaluation procedure was selected for the micrographs obtained in run 1. Here, because of the relatively weak overall deformation,

only a small number of grains in the  $\alpha$  phase had recrystallized. In this case, the total area capable of recrystallizing (as determined by heating into the grain-coarsening range at the end of the run), and not the total area of the  $\alpha$  phase, was taken as 100% and the fraction of the saturation values calculated on this basis. The smallest overall reduction required to cause the entire  $\alpha$  phase to recrystallize was found to lie between 7.7 and 11.3%.

If one plots the saturation values obtained for a given reduction in area against recrystallization temperature, it is found that the best regression curve through the calculated areas is the straight line. This is shown in Figs. 3 and 4. On either side of the test points the temperature variation at  $T_c$  is indicated. For small deformations only two points were evaluated quantitatively since the temperature band of the recrystallization range is very narrow. The corresponding graph for pure copper is shown in Fig. 5. In this case too the best fit is the straight line.

### Onset of Recrystallization

At both  $T_{c1}$  and  $T_{c2}$ , a certain delay time precedes the onset or the continuation of recrystallization. For instance, it can be seen from Fig. 1 that  $\sim 40$  min elapse, after the temperature has been raised from 415 to  $420^\circ\text{C}$ , before an increase in recrystallized areas materializes. For the three runs (5, 6, and 7) of the samples deformed 21.9%, we determined the delay time preceding recrystallization at  $T_{c1}$  ( $t_B$ ) and the time necessary to reach saturation at  $T_{c1}$  ( $t_s$ ). In view of the fact that pictures were taken in the PEEM only every half hour, the values for  $t_B$  and  $t_s$  are rather imprecise. Yet, when the reciprocal values for these two parameters are plotted on a logarithmic scale against temperature (Fig. 6) one finds that the points lie on straight lines that cut each other at  $\sim 406^\circ\text{C}$ . Although these two lines were based on an absolute minimum of points, it is interesting to observe that they nevertheless lead to exactly the same minimum recrystallization temperature as can be found by extrapolation from the corresponding curve in Fig. 4. Moreover, the lines in Fig. 6 suggest the following tendencies: at low recrystallization temperatures the delay time preceding the onset of recrystallization is long and the time interval to reach saturation short; at high recrystallization temperatures these tendencies are reversed.

### The Limits of the Recrystallization Range

From the regression lines in Figs. 3 and 4 it is possible to determine by extrapolation the lower and the upper limit of the temperature band within which partial recrystallization occurs. The temperatures for 0 and for 100% recrystallization, respectively, are plotted against reduction in area in Fig. 7.



Fig. 2 (continued on p. 28)

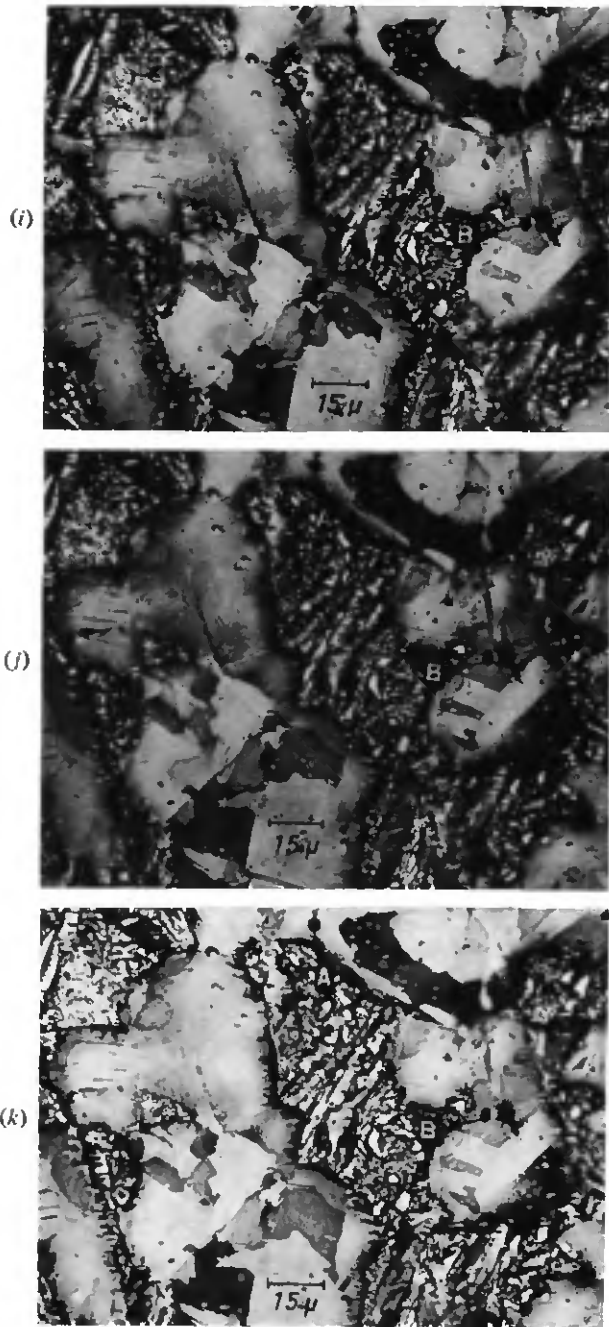


Fig. 2(a)–(k) Photomicrographs taken in the PEEM during heating run 3 (Cu–10% Al, 17.1% prior deformation). (a)–(f) were taken at 415°C, (g)–(k) were taken at 420°C, at the times indicated. Points A and B indicate the area where the first recrystallized grains appeared.

It may be noted that the lower limit decreases with increasing prior deformation, whereas the upper limit increases rapidly from ~ 11.3% onwards to reach a limiting value at ~ 20% reduction in area. It is evident, too, that the temperature range of partial recrystallization becomes greater with deformation.

Fig. 6 Reciprocal values of the incubation time for recrystallization ( $t_B$ ) and of the time necessary to reach saturation ( $t_s$ ) at different  $T_{01}$  temperatures. (Cu–10% Al, 21.9% prior deformation.)

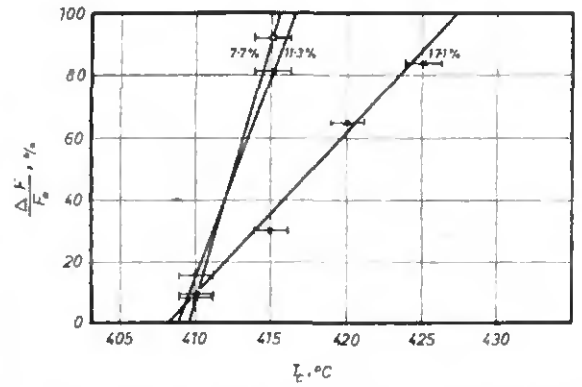


Fig. 3 Saturation values plotted vs. recrystallization temperature for three different amounts of prior deformation. (Cu–10% Al.)

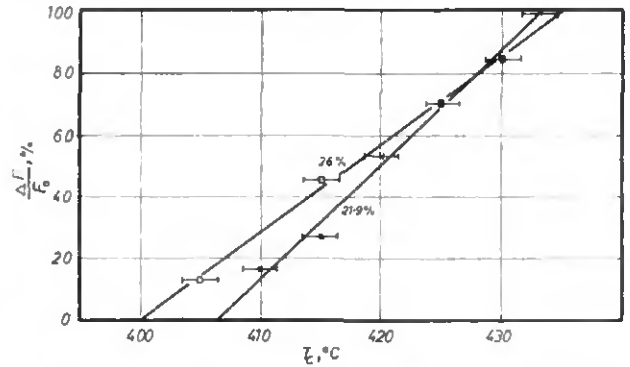


Fig. 4 Saturation values plotted vs. recrystallization temperature for two different amounts of prior deformation. (Cu–10% Al.)

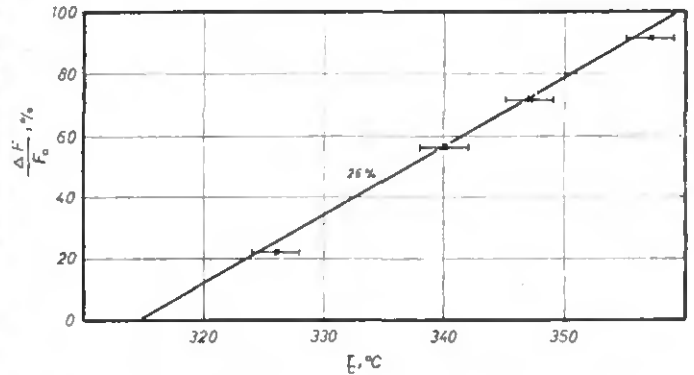
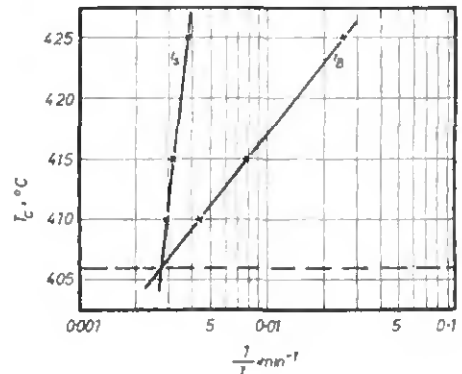


Fig. 5 Saturation values plotted vs. recrystallization temperature for pure copper deformed 26%.



### Effect of Grain Boundaries in the Deformed Matrix

From the pictures taken in the PEEM an attempt was made to determine to what extent grain boundaries in the deformed matrix were able to act as preferred sites for recrystallization initiation. For this purpose we assessed the frequency with which new grains appeared at the boundaries relative to that with which such grains appeared in the interior of as-deformed grains. It was appreciated that the appearance of a new grain in the interior of a deformed grain did not necessarily mean that it had nucleated there; it could well have been formed at an old boundary, but below the surface of observation and growing at an angle to the latter. Despite this possibility it was found, however, that three times as many new crystals formed at a boundary of the original grain structure as in the grain interior, thus emphasizing the predominant role played by the grain boundaries of the deformed matrix.

Furthermore, it was found that the frequency with which new crystals appeared at the  $\alpha/\alpha$ , the  $\alpha/\text{eutectoid}$ , and the twin boundaries, respectively, was in the ratio 23 : 6 : 4, thus attributing the major role in providing preferred nucleation sites clearly to  $\alpha/\alpha$  boundaries.

We should also like to emphasize in this connection that in no instance did we observe the bowing out of an old grain boundary to initiate the formation of a new grain, as proposed in the nucleation model of Bailey and Hirsch.<sup>6</sup> As a matter of fact, the deformed matrix must be considered as an 'immobile mass', which participates in the recrystallization process only in the sense that it furnishes the material for the new crystalline body formed as a result of recrystallization.

### The Meaning of Grain Size

The two micrographs in Fig. 8, each taken close to completion of the recrystallization process, show that the average recrystallized grain size after severe deformation seems to be smaller than in a previously lightly deformed matrix. This is a well-known fact. It is even widely held that the average grain size becomes larger with increasing recrystallization temperature under the slow heating rates employed here. Yet, since in the domain of recrystallization under consideration, i.e. in that temperature range where partial recrystallization occurs, one always finds a mixture of deformed and of recrystallizing grains, it is meaningless to indicate an average recrystallized grain size. A clear-cut grain-size determination can obviously be made only at the temperature for which the saturation value  $S = 100\%$ . Beyond this temperature one must expect grain growth to occur sooner or later, leading to a mixture of recrystallized grains and of those that have already coarsened at the expense of others.

Finally, we should like to point out that one of the uncertainties relating to our quantitative measurements stems from the fact that we observe the surface and not the interior of our samples, and that what happens at a free surface may not be representative of what happens inside a body. This point was checked by successive removal of material and intermittent observation in the PEEM of a specimen containing large grains and having recrystallized by only 10%. These two latter conditions permit a distinction to be made between the deformed and the recrystallized part of the surface, without requiring continuous observation to achieve this as is necessary for more advanced recrystallization stages or smaller grain sizes.

No significant difference was found between the originally determined value of the recrystallized area and that measured and calculated after a total removal of 1 mm. Thus, one can make a direct determination of the recrystallized volume when the recrystallized area is known.

### Discussion

#### General

The results presented here show that a temperature range does indeed exist within which recrystallization proceeds only partially and within which the Avrami-type equation cannot describe the recrystallization kinetics.

The main reason why this has been unrecognized for such a long time must be sought above all in the fact that until now—except in special cases—the deformed grains, the recrystallizing grains, and the grains in the process of coarsening could not readily be distinguished from one another by those workers who attempted to study recrystallization by direct observation. That this difficulty is a very real one can easily be seen from Fig. 9, obtained with a copper specimen previously deformed by 26%. It is absolutely impossible to deduce from the PEEM picture in Fig. 9(a) alone that 56% of the sample has already recrystallized. The same applies to the picture taken in an optical microscope after cooling the same sample down to room temperature and removing it from the PEEM (Fig. 9(b)). To be able to distinguish clearly between the old and the new structure one has to examine a sequence of pictures belonging to the same isothermal run.

To be sure, recrystallization also takes place above that temperature ( $T_{100\%}$ ) which must be attained for recrystallization to go to completion. Yet, when this temperature is exceeded, grain growth, which obeys an entirely different kinetic law, is likely to occur.

It should, therefore, be evident that, when the various states in question (deformed, recrystallized, grain-coarsened) are mixed up, it is impossible to arrive at an unambiguous analysis of the recrystallization process. Judging from the general acceptance of Avrami's equation, one is led to the belief that most of the researchers who studied recrystallization by discontinuous observation of the structure of a number of specimens have in fact worked above the actual recrystallization range, i.e. most probably in the temperature range of grain growth. Such an assumption is also confirmed by the indication<sup>19</sup> that the grain size during recrystallization is time-dependent. Since for ordinary grain sizes (10–50  $\mu\text{m}$ ) the driving force for grain growth is  $< 1\%$  of that operating during recrystallization, it follows that the respective studies yielded results that cannot be interpreted unambiguously.

One of the vital aspects of the present work is, thus, that it was possible to make a clear-cut distinction between the various stages in the sequence of deformed and annealed structures. In view of our results we can now define the basic range of recrystallization as follows:

For a given initial grain size and specified deformation a temperature range exists within which the formation of new, strain-free, and largely equiaxed grains proceeds only partially. At each temperature recrystallization reaches a limiting value, termed 'saturation'. The highest temperature for which the saturation value is zero is the 'lower (or minimum)' recrystallization temperature, whereas that temperature at which saturation just attains 100% is termed the 'upper' recrystallization temperature. The latter is also the minimum temperature at which recrystallization goes to completion without being accompanied by grain-coarsening.

#### The Saturation Phenomenon

The fact that within a given temperature range recrystallization does not go to completion, even if a specimen is held at  $T_c$  for an indefinite period of time, is contrary to all theories of recrystallization so far advanced.

Since nuclei are always limited in number, it follows that the cause of partial recrystallization cannot be associated with

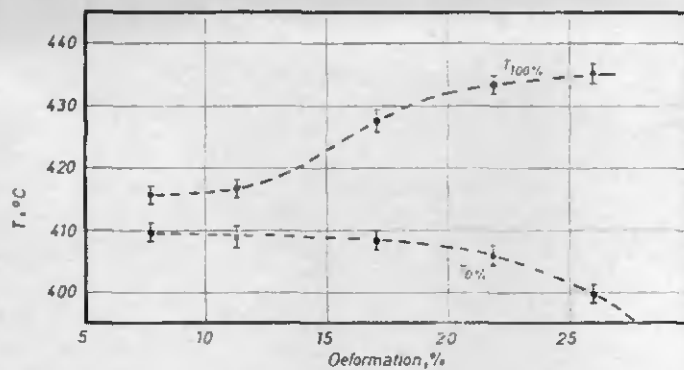


Fig. 7 The lower ( $T_{0\%}$ ) and the upper ( $T_{100\%}$ ) recrystallization temperature as a function of prior deformation. (Cu-10% Al.)

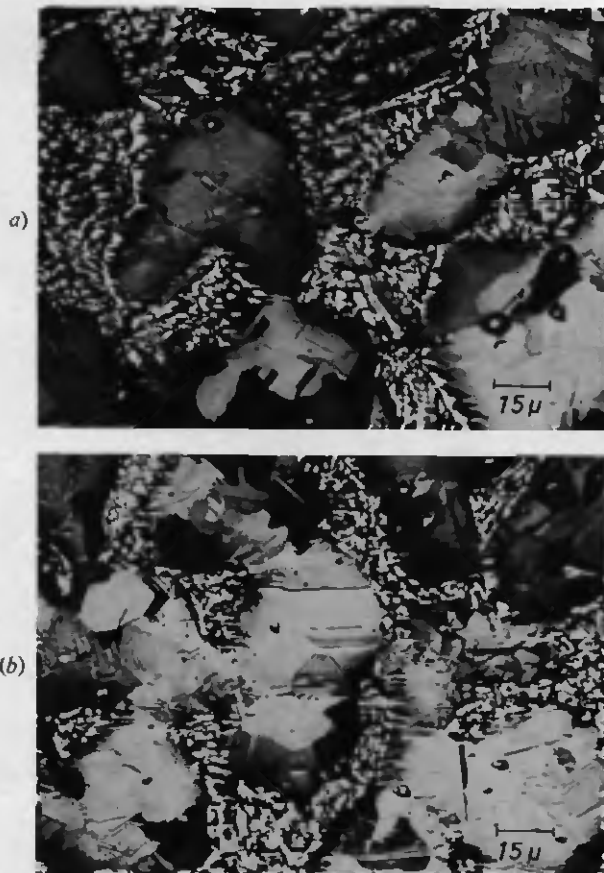


Fig. 8 Photomicrographs of Cu-10% Al taken close to completion of recrystallization, illustrating the effect of prior deformation on recrystallized grain size. (a) Deformed 11.3%,  $T_c = 415^\circ\text{C}$ ,  $S = 81.6\%$ ; (b) deformed 26%,  $T_c = 435^\circ\text{C}$ ,  $S = 84.8\%$ .

nucleation but must be sought in the blocking of the interface movement of the newly formed grains. If such an impediment were not operative, then those nuclei that become active at the lowest recrystallization temperature would suffice to reach 100% recrystallization at that temperature, as is generally believed to be the case. Yet, as pointed out above, this is not what actually happens. Furthermore, in the absence of a blocking mechanism acting against the interface, one would also expect the recrystallized grain to become finer as the recrystallization temperature was increased, since more nuclei become active as the temperature is raised. One may

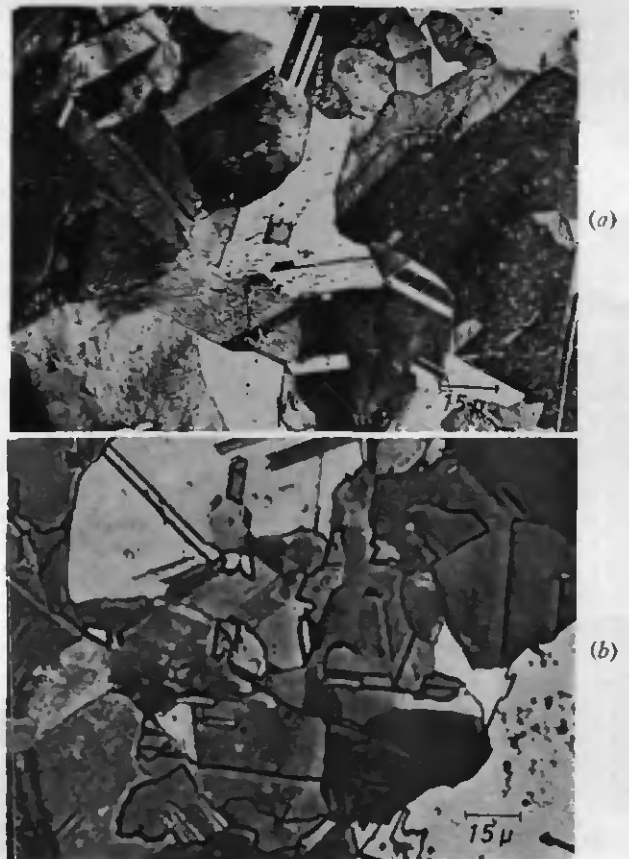


Fig. 9 Comparison between a photomicrograph taken (a) in the PEEM, and (b) subsequently under the optical microscope (not at exactly the same position!) of a sample which had already recrystallized by 56%. (ofhc Cu, 26% prior deformation.)

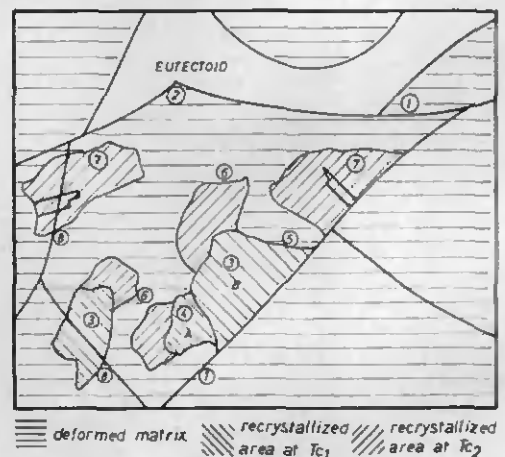


Fig. 10 Schematic drawing showing a number of possible means by which the recrystallized area increases as a function of time and temperature:

- (1) Deformed  $\alpha/\alpha$  boundary.
- (2) Boundary between  $\alpha$  and eutectoid.

Changes at  $T_{c1}$ :

- (3) New grain nucleated at  $T_{c1}$ .
- (4) New grain (A) nucleated at  $T_{c1}$  on a new, but previously nucleated, grain (B) with an orientation different from that of A.

Changes at  $T_{c2}$ :

- (5) Boundary blocked at  $T_{c2}$ .
- (6) Moving boundary that nucleated at  $T_{c1}$ .
- (7) New grain nucleated at  $T_{c2}$ .
- (8) Shows movement of new grains across an old boundary of the deformed matrix.

even go further and propose that the same final grain size could be obtained by recrystallizing a highly deformed material at a low temperature as for a lightly deformed material recrystallized at a high temperature. Clearly, this has never been observed under the slow heating rates employed here and in conventional annealing practice.

It is difficult, with the evidence at hand, to determine the precise nature of the blocking mechanism that is supposed to produce the saturation effect. First, despite all claims to the contrary, one does not really know what a recrystallization nucleus looks like; moreover, one is also ignorant about the precise mechanism by which a moving boundary sweeps up the dislocations. Nevertheless, the following possibilities may be envisaged:

(1) *The moving boundary is blocked by impurities.*

Here we should like to repeat that in no instance did we observe the movement of a boundary that existed in the deformed matrix. Therefore, if these were blocked, this should not have an effect on newly formed grain boundaries. Moreover, the saturation effect is also typical for the recrystallization of pure copper. In this latter case the gradient of the straight line (Fig. 5) is even appreciably shallower than for the Cu-10% Al alloy deformed by the same amount. On the basis that the aluminium bronze is considerably less pure than the Cu (ofhc), it may be concluded that the saturation effect cannot be associated with anchoring of the newly formed boundaries by impurities. For analogous reasons one can also exclude the eutectoid phase as playing a governing role in the saturation phenomenon.

(2) *An equilibrium is established between the driving force of recrystallization (energy associated with the difference in dislocation density) and the increase in surface energy required for the formation of the new, recrystallized grains.*

If this were the case, one would expect that the saturation value at a given  $T_c$  would be highest for a heavily deformed material, since the large driving force could provide for a considerable amount of new surface energy. However, a look at Figs. 3 and 4 confirms exactly the opposite tendency.

(3) *The new grain boundaries are blocked by special dislocation configurations.*

One could imagine that a grain boundary must lie at a given angle (or within a certain range of angles) relative to the dislocation arrangement ahead of it, for it to be able to sweep up the latter.

The mechanism actually operating that accounts for the saturation effect requires observation in the TEM, since only in this fashion can one hope to detect the reasons why, at a given temperature, the capability of a grain boundary to sweep up dislocations is limited.

#### The Limits of the Recrystallization Range

The variation in the lower recrystallization temperature with deformation, as illustrated in Fig. 7, is what one intuitively expects on the basis that a higher dislocation density facilitates nucleation of new, strain-free grains.

As far as the upper limit of the recrystallization range,  $T_{100\%}$ , and its variation with prior deformation are concerned, this cannot be understood on the basis of present-day ideas about recrystallization. All the arguments that could be advanced in the light of such concepts lead to a decrease in  $T_{100\%}$  with increasing deformation; in fact, one would reason that the increase in dislocation density accompanying an increase in prior deformation would facilitate both nucleation

and movement of newly formed boundaries. Yet the results suggest that recrystallization becomes more difficult with increasing deformation, as if the tangled state of dislocations might hinder their being swept up by a moving grain boundary.

In this connection it must be pointed out that when measuring the increase in recrystallized area—whether as a function of time or when passing from  $T_{c1}$  to  $T_{c2}$ —no distinction was made between newly nucleated grains and those that simply continued to grow before impinging upon one another. Fig. 10 shows the various possible means by which the recrystallized area can increase. Thus, what was plotted in Figs. 3-5 is the effect resulting both from the nucleation of new grains and the extension of non-impinged but already existing, grains. An interpretation of the underlying mechanism would require a separation of these two contributions. This is being carried out as a further phase of the present work.

Finally, it is also interesting to note that the straight lines from which  $T_{0\%}$  and  $T_{100\%}$  were found by extrapolation, were not obtained from observations on one sample only, but from two or three specimens (see Table I) depending on the amount of preceding deformation. This suggests either that the samples were large enough to guarantee a valid statistical mean value or that they were taken from a very homogeneous material, or both.

#### Model of a Possible Recrystallization Mechanism

On the assumption:

(i) that a grain boundary at a given temperature has a limited and specific capability to sweep up dislocations,

(ii) that this capability increases with temperature, whereby the point defects may play a role in the rearrangement of the dislocations, one may try to explain the observed phenomena reported here in the following manner.

In the case of small deformations (and thus low densities and relatively simple arrangements of dislocations) the capability to sweep up dislocations of a small number of grains suffices to recrystallize the entire matrix. A small number of relatively large grains is thus obtained over a narrow temperature band. After larger deformations the sweeping-up capacity of the grain boundaries is exhausted more rapidly, and this probably not only because of the higher density but also because of the more complex tangles of dislocations. For recrystallization to continue, the temperature must be raised. This increases the sweeping-up capability of the boundaries and, at the same time nucleates new grains. From the shape of the  $T_{100\%}$  curve and the rather uniform new grain size throughout the partial recrystallization range, one may conclude that, particularly after severe deformation, it is easier to increase the total grain-boundary area by the nucleation of additional grains when raising the temperature than by the continued movement of boundaries of grains already recrystallized at a lower temperature.

The above proposition does not include any suggestion as to how the sweeping-up of dislocations in a grain boundary occurs.

#### Summary

Thanks to the availability of a photoemission electron microscope it has been possible to demonstrate the existence of a temperature range within which recrystallization proceeds only partially, irrespective of how long a specimen is held at a temperature within this range. It could thus be shown that an Avrami-type equation cannot be used to describe the basic recrystallization kinetics.

By plotting the limiting (saturation) value reached at a given temperature vs. temperature a straight line is obtained which, by extrapolation, permits a clear and unambiguous definition of the lower and the upper recrystallization temperature for each prior deformation.

The greater the deformation the easier it is to form the first recrystallizing grain, but the more difficult it is to complete recrystallization.

Since in the recrystallization range deformed grains and recrystallizing grains coexist, it is of no value in that connection to speak about an average grain size. Rather, a clear-cut

grain-size determination can be made only for the temperature,  $T_{100\%}$ , at which recrystallization goes to completion, or eventually at a higher level, provided that grain coarsening does not occur.

#### Acknowledgements

The authors gratefully acknowledge the financial support of the Entwicklungsfonds für seltene Metalle and the Fonds National Suisse de Recherche Scientifique, without which the work reported here could not have been carried out.

#### References

1. R. F. Mehl, 'Physical Metallurgy' (edited by R. W. Cahn), p. 10. 1970: Amsterdam (North Holland).
2. M. Avrami, *J. Chem. Phys.*, 1939, 7, 1103.
3. W. Johnson and R. F. Mehl, *Trans. Amer. Inst. Min. Met. Eng.*, 1939, 135, 416.
4. F. Haessner, 'Recrystallization of Metallic Materials', p. 5. 1971: Stuttgart (Riederer Verlag).
5. R. W. Cahn, 'Recrystallization, Grain Growth, and Textures', p. 99. 1966: Metals Park, Ohio (Amer. Soc. Metals).
6. J. E. Bailey and P. B. Hirsch, *Proc. Roy. Soc.*, 1962, [A], 267, 11.
7. J. W. Christian, 'The Theory of Transformations in Metals and Alloys', p. 718. 1965: Oxford, &c. (Pergamon Press).
8. P. Gordon and R. A. Vandermeer, Ref. 5, p. 205.
9. J. K. Stanley and R. F. Mehl, *Trans. Amer. Inst. Min. Met. Eng.*, 1942, 150, 260.
10. L. Wegmann, 'Photoemissions-Elektronenmikroskopie', 'Handbuch der zerstörungsfreien Materialprüfung'. 1969: München (R. Oldenbourg).
11. M. S. Schweizer and G. W. Form, *Metals and Materials*, 1970, 4, 369.
12. G. Gindraux and G. W. Form, *Mém. Sci. Rev. Mét.*, 1970, 67, 617.
13. S. Murphy and C. J. Ball, *J. Inst. Metals*, 1972, 100, 225.
14. Ref. 4, pp. 9, 109.
15. R. W. K. Honeycombe, 'The Plastic Deformation of Metals', p. 301. 1968: London (Edward Arnold).
16. 'Recovery and Recrystallization of Metals' (edited by L. Himmel), p. 103. 1963: New York and London (Interscience).
17. G. R. Spicich and R. M. Fisher, Ref. 5, p. 563.
18. M. Hansen and K. Anderko, 'Constitution of Binary Alloys', p. 85. 1958: New York and London (McGraw-Hill).
19. W. Charnock and J. Nutting, *Metal Sci. J.*, 1967, 1, 78.

© The Institute of Metals. 1973.

Cette publication résume la thèse de doctorat soutenue à l'Université de Neuchâtel par M. Maurizio Schweizer le 5 juillet 1972.