

Physics. — *Recovery and recrystallization viewed as processes of dissolution and movement of dislocations. I.* By W. G. BURGERS. (Laboratorium voor Physische Scheikunde der Technische Hoogeschool, Delft.) (Communicated by Prof. J. M. BURGERS.)

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Summary.

In this paper recovery and recrystallization are treated as processes of dissolution and movement of dislocations present in the cold-worked state.

In I some general features of both processes are discussed, especially with a view to ascertain whether they are fundamentally different or whether recovery has to be viewed as recrystallization on an "invisible" scale. Some experimental facts are brought forward, relating to the dependence of both phenomena on time and temperature of annealing and degree of deformation, which point to fundamental differences between both processes.

In order to get a closer insight into these differences, in II the block-structure of the crystalline state is discussed. Attention is drawn to the fact that single crystal, polycrystal and cold-worked state differ only by degrees: they all are built up of lattice blocks, eventually in a stressed condition, connected and separated by transition layers of dislocated atoms. Some considerations regarding the stability of such structures, as far as available from the literature, are given.

Finally in III the atomic movements, induced in the cold-worked state by heating, are considered. A difference is made between (a) displacements which, by "dissolution" of dislocations, change the stresses in the lattice-blocks, and (b) displacements of boundary layers as a whole, causing growth of one domain at the cost of an adjoining one. The former are essential for recovery, the latter for recrystallization. With the aid of both types of displacements several experimental facts, observed with recovery and recrystallization, are discussed. As to recrystallization it is mentioned that the mean rate of growth of crystals, grown in fine grained quasi-isotropic polycrystalline testpieces, although being constant in all directions, is different for different crystals. The possibility is to be considered that such differences are due to differences in imperfection ("mosaic character") of these crystals. This supposition would imply the assumption that a crystal, when starting its growth from an "imperfect" nucleus, retains this imperfection throughout its development.

Finally the occurrence of "stimulated" crystals is brought forward. It is shown that their presence explains the increase in rate of nucleation in the course of isothermal recrystallization, observed recently in an investigation of the recrystallization of aluminium by ANDERSON and MEHL.

Introduction.

When a metal is cold-worked, its physical and mechanical properties generally suffer considerable changes. By a subsequent annealing treatment these changes are again annihilated, either gradually or, at a sufficiently high temperature, in a more or less abrupt manner. We can thus speak of a process of recovery, taking this word in its general sense. In practice, however, there is reason to discriminate between two apparently different processes, one most easily observed at lower temperatures, in which a return of the properties to their normal values takes place without "visible" change in structure of the metal, and a second process, in which we observe the formation and the growth of new crystallites. The expression *recovery* is generally confined to the first mentioned process, whereas the second is called *recrystallization*. This expression embraces in its "primary" stage the formation and growth of new crystallites in the deformed matrix. It is often followed by a "secondary" growth of the new crystallites at the cost of their equally new neighbours, a process indicated as grain-growth.

Although both recovery and recrystallization have been extensively studied, for example in their dependence on duration and temperature of annealing (for a summarizing review we may refer to (1)), it must be stated that our knowledge and insight in the mechanism of the underlying atomic processes are still very defective, notwithstanding valuable contributions to this question by various authors, of whom we mention in particular KORNFIELD (2) and DEHLINGER (3).

In what follows we intend to discuss recovery and recrystallization, starting from a picture of the cold-worked state as consisting of a system of slightly deformed lattice blocks, separated and connected by more severely dislocated transition layers. One of our objects will be to ascertain in how far such a picture can be of help to understand some of the differences observed between these two annealing processes.

I. Recovery versus recrystallization in their dependence on time and temperature of heating.

I, 1. Analogy.

The question can be raised what, physically speaking, the difference between recovery and recrystallization amounts to, as undoubtedly both processes are due to the shifting of a certain number of atoms in the deformed matrix to positions of more stable equilibrium. In fact it is often very difficult to decide whether during a given heat-treatment recrystallization does not occur at all. The dissolving power of the experimental method applied to ascertain the presence or absence of structural changes (microscopic observation, X-ray diffraction, etc.) plays an important part here. Often what is called recovery according to one method, is beginning recrystallization according to the other.

In practice a discrimination between recovery and recrystallization is

certainly justified. Especially at lower temperatures of heating a considerable recovery of physical and mechanical properties can be brought about without "visible" structural changes. This is particularly true with deformed *single* crystals, where, according to measurements of HAASE and SCHMID (4) with bismuth and zinc, of KORNFELD (5) with aluminium and of KOREF (6) with tungsten, an approximately complete recovery of the shear- or tensile-stress could be brought about. With polycrystalline testpieces such a "pure" recovery cannot be realized so completely.

On the other hand it was brought forward in particular by VAN LIEMPT (7), that in many annealing experiments the relation between time (t) and absolute temperature (T) of heating, required to cause a definite percentage of recovery of a physical property (for example the electric resistance) or in other cases a definite state of recrystallization (for example half of the matrix recrystallized) obeys to the same formula, of type

$$T \ln c \cdot t = \frac{Q}{R} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

in which Q has the meaning of an activation energy for atomic interchange and c is proportional to the atomic frequency of the metal considered.

This "law of corresponding states of recovery (or recrystallization)" may said to be based on the very simplest picture of the processes involved, in which the atomic movements from a metastable "deformed" into a stable "undeformed" position are governed by a single activation energy Q . In such a picture the recovered respectively recrystallized fraction (x)¹⁾ at any moment obeys a relation

$$\frac{dx}{dt} = k(1 - x) \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

with the velocity constant k determined by

$$k = c \cdot e^{-\frac{Q}{RT}} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

or integrated

$$x = 1 - e^{-c \cdot t \cdot e^{-Q/RT}} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

[see KRUPKOWSKI and BALICKI (8); also (1) (§§ 75, 77, 151)]. For the quantity Q values are found of the order of magnitude of those met with in diffusion experiments in the solid state, that is, values to be expected for processes, consisting of interchange of individual atoms.

Similar activation energies occur in the relations, which in "pure" recrystallization experiments are found to govern the rate of nucleation and the rate of growth of the new crystals, for example with aluminium by

¹⁾ It is further assumed that the relative recovery of the property considered is at every moment proportional to this fraction.

KORNFELD and PAWLOW (9) and by ANDERSON and MEHL (10), for rock-salt by MÜLLER (11) ²⁾.

This apparently closely analogous behaviour of both recovery and recrystallization in their dependence on time and temperature of heating might be considered to support the conception, that what is called recovery is in reality recrystallization on such a small scale, that it is still "invisible", so that there would be no reason to discriminate between both phenomena in a more fundamental way.

I, 2. Differences.

This conclusion breaks down, however, if we consider what happens after *prolonged times of heating*. If, under such circumstances, "visible" recrystallization sets in, we observe a return of the properties to their normal values for the completely annealed state, this state being attained the sooner the higher the chosen temperature, as might be expected after [1], which is confirmed in the more precise treatment given in (10). In "pure" recovery-anneals, on the contrary, after an initial return of the properties, the rate of the process decreases so much below the magnitude

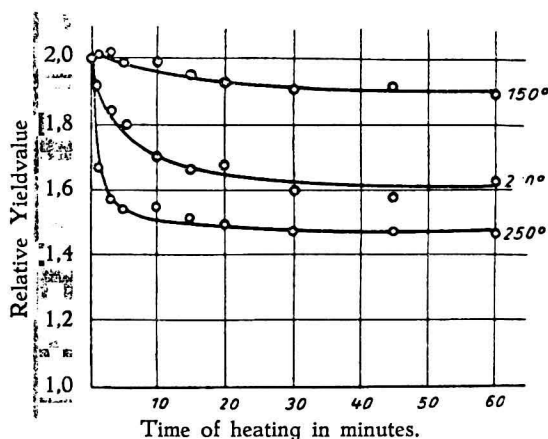


Fig. 1. Isothermal recovery curves of aluminium single crystals. The original deformation was in all cases the same (100% increase of yield value = relative yield value 2). The ordinates give the decrease of this latter quantity as a function of time of heating for three different temperatures: *at each temperature a nearly constant "rest value" is approached, which lies the lower the higher the temperature of annealing (after KORNFELD (2) (5)).*

²⁾ In ANDERSON and MEHL's paper, which appeared as recently as 1945, the course of recrystallization in aluminium sheet was analysed in terms of rate of nucleation (N = number of new crystals formed per unit of time per unit of area of the unrecrystallized matrix) and rate of growth (G). It follows from their investigation that formula [1] given above is valid only under simplifying assumptions (for that case the paper gives a relation between Q and the activation energies Q_N and Q_G for nucleation and growth separately). ANDERSON and MEHL show that the whole course of isothermal recrystallization is more complicated than follows from [2] and [3], in particular in consequence of the circumstance, that N increases with time, a fact to which we shall return at the end of section III of this paper.

expected according to [4], that a value is attained which remains approximately constant for much longer durations of heating. Although the difference between this "restvalue" and the value for the completely annealed state diminishes with increasing temperature of annealing, nevertheless at each temperature, if not too low, a definite residual effect of the cold-working remains. Fig. 1, which is taken from KORNFIELD (2) (5), may serve to illustrate this for annealed aluminium crystals [for other examples we refer to (1) (§ 69)].

This different behaviour of "recovering" and "recrystallizing" anneals is still more accentuated by the fact that, according to KORNFIELD (2), the rate at which the "restvalues" are attained, is greater for single crystals than for polycrystals: this is contrary to what is generally found in recrystallization experiments: here for a given deformation polycrystalline material recrystallizes "easier" (for example at lower temperatures) than unicastalline material [KARNOP and SACHS (11a); (1) (§ 150)].

A difference is also observed when we consider annealing experiments carried out with test-pieces, subjected to *different degrees of cold work*. On general grounds one would expect the activation energy Q to decrease with increasing deformation and thus, according to [4], the more severely deformed test-piece to approach the completely annealed state for a given temperature in a shorter time, or for a given time at a lower temperature. This is actually observed in pure recrystallization experiments

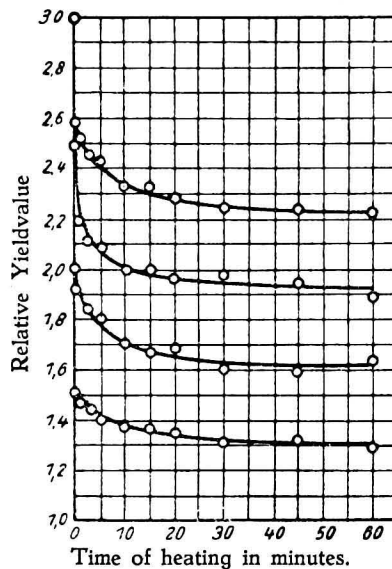


Fig. 2. As fig. 1. Here, however, all curves relate to the same temperature (200° C.) but to four different states of deformation, the original relative yield values before annealing being 3, 2.5, 2 and 1.5. The "rest values" lie higher, the larger the original degree of cold-work (the relative recovery¹⁾ is approximately the same in all four cases, namely 0.4) (after KORNFIELD (5)).

¹⁾ Calculated as:
$$\frac{\text{yieldvalue before anneal} - \text{yieldvalue after anneal}}{\text{yieldvalue before anneal} - \text{original yieldvalue}}$$

[see again ANDERSON and MEHL (10)] and also in such "recovery" experiments, in which it is highly probable that the recovery was actually accompanied by recrystallization: for example by SAUERWALD (12), BRINDLEY (12a). In KORNFIELD's (5) recovery experiments with aluminium single crystals, however, a different result is obtained, as shown in fig. 2. Here the four "recovery isotherms" relate to four crystals, extended so far that the yield-value was raised to respectively 1.5, 2, 2.5 and 3 times its original value. The curves show the decrease of these "relative" elastic limits as a function of the time of heating at 200° C., where no "visible" recrystallization occurred. It will be seen that the quantities considered approach to residual values which lie *higher the more the crystal had been deformed* (the *relative* recovery, however, was approximately the same for all degrees of deformation). This result again is quite different from that generally found in the case of recrystallizing test-pieces³⁾. MASING (12b) has already drawn attention to this curious behaviour and considered it as an indication that recovery and recrystallization are essentially different phenomena⁴⁾.

Leaving this question as it stands, it seems in any case to follow from what has been said that "pure" recovery at constant temperature of annealing leads to states, which are intermediate between the cold-worked and the completely annealed state and which possess a considerable stability.

³⁾ A to some extent similar result was, however, obtained by BRINDLEY (12a) in annealing experiments with rolled copper with different percentage reduction, where the thermal E.M.F. between the cold-worked and the undeformed state was measured, although in these experiments the occurrence of at least partial recrystallization seems possible. According to BRINDLEY it is likely, that the effect is due to different grain sizes and (or) grain orientations in the partly annealed states as compared to those present in the original undeformed metal.

⁴⁾ It must be remarked here that also DEHLINGER (3) has advanced the view that recovery and recrystallization are different processes. To his conceptions we shall return in section III. DEHLINGER's arguments were partly based on the assumption of an essentially different dependence of both phenomena on temperature, in the sense that the temperature coefficient of the velocity for recrystallization was much larger than that for recovery. As far back as 1929 DEHLINGER in a fundamental paper on recrystallization put forward the conception of an "instability-temperature" (a kind of CURIE-temperature), above which the rate of recrystallization should increase jumpingly. As we exposed in (1), (§ 152), we do not know experiments which support this assumption in an unambiguous way; also its theoretical basis is not clear.