The Effect of Sudden Heating on the Recrystallization of Metals

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Abstract

In recrystallizing thin aluminium plates the effect of sudden heating to a recrystallization temperature belonging to the germinative temperature range upon the grain number was investigated, and it was found that the grain number is much greater in this case than when the plate is heated gradually up to the same recrystallization temperature. In the case of gradual heating, a few seeds or crystal nuclei which are to be grown to tolerable size afterwards in the germinative temperature range seem to be formed among an immense number of the recrystallized metal crystals already in the recrystallization temperature range lower than the germinative temperature.

Recently one of the writers and Koyanagi[1] published an opinion on the recrystallization of metals. In that occasion they assumed that the melting point of a more distorted crystal is lower than that of a less distorted one, and that the metal is easier to melt at its crystal boundaries than in the interior of individual crystals; and the process of recrystallization was considered to be the growth of almost undistorted perfect crystal nuclei formed in the melts at the crystal boundaries. This theory is of course a rough one, and must be subjected to closer examination from various standpoints concerning this phenomenon, which may request some refinement or modification at least. Following this purpose the writers began by examining the effect of sudden heating on the recrystallization of thin aluminium plate in the present experiment.

In order to ensure as far as possible a simultaneous sudden heating of the whole portion of the specimen, thin commercial aluminium plate of 1 m.m. thickness was used throughout the present experiment; and small test pieces having the areal size of 10 c.m. × 1.5 c.m. were cut out from the plate. The furnace used in the present experiment was a resistance type, consisting of a porcelain tube about one metre in length and five centimetres in diameter and a nichrome wire wound uniformly around it. The furnace was set vertically, and the lower end of the porcelain tube was plugged tightly with asbestos.

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1. U. Yoshida and K. Koyanagi: These Memoirs, **18**, 9 (1935)

At the upper end of the tube a cover of asbestos plate is provided which facilitates the operation of opening and covering the upper end of the tube. In exposing a test piece to some high temperature, it was always hung free at the middle of the furnace by means of a wire hooked at the upper end of the porcelain tube; and the temperature to which the test pieces were exposed was measured by a thermocouple, whose one junction was placed at the middle part of the furnace. In the case of exposing a test piece to some required high temperature we must at first properly regulate the heating electric current through the nichrome wire of the furnace, so that the required high temperature is maintained steadily for a long time. After such a temperature is established in the furnace, we insert the test piece into the furnace as quickly as possible in the manner stated above by opening and covering the asbestos cover. This process can be done very quickly within the time of two or three seconds; and the resulting slight lowering of the temperature of the furnace caused by this process is recovered automatically to the initial steady temperature within several seconds.

In the present experiment all the test pieces of commercial aluminium plate were subjected to a preliminary annealing for 24 hours at the temperature of the furnace of 320°C; then they were elongated at room temperature, for various amounts of elongation ranging from 1 percent to 10 percent of the original length. Such cold-worked test pieces were next exposed suddenly, unless otherwise described, to various definite recrystallization temperatures one by one in the manner stated before; and the aspect of the test pieces thus recrystallized was revealed to the naked eye by etching the surface of the test pieces with some reagent after they were taken out of the furnace.

First of all the writers examined the relation between the recrystalization temperature, the amount of the cold-working of the test piece before recrystallization, and the number of the crystals formed by recrystallization, which were contained in the unit areas of the surface of the test pieces. The results obtained are tabulated in Table I. For the cold-working the writers elongated the specimens by stretching, and the percentage of elongation of the original length was taken as the amount of cold working, as is given in the table. Here it must be noted that, though the time of recrystallization which is needed to complete the grain growth is different according to the amount of the cold working and the recrystallization temperature, it was taken for a time long enough that the grain growth was ended and the grain boundaries became entirely sharp. The counting of the crystal number was made at six different portions of each specimen, and the average number per square centimetre is given in the table.

Table I

Number of grains/cm², (sudden heating)

Recrystallization temperature \ Elongation in %	10	7	6	3	2
645°C	145	64	43	31	31
630°	49	54	39	28	27
600°	114	47	40	18	17
550°	27	38	11	6	—
500°	90	15	3	—	—

As is seen in Table I, the grain number per unit area increases, for a given recrystallization temperature, with increased amount of cold working, as is usually the case with gradual heating; and it increases, for a given amount of cold working, with increased recrystallization temperature. This temperature relation of the grain number is entirely different from the case of gradual heating, in which case the grain number per unit area does not increase, but rather tends to decrease with increased recrystallization temperature due to the growth of the crystals. The grain number per unit area is much greater in the case of sudden heating than when the temperature of the furnace is raised gradually to the maximum recrystallization temperature. To make clear this point, another recrystallization experiment with the test pieces having the same size as above was carried out by raising the temperature of the furnace gradually from the room temperature to the maximum recrystallization temperature of 640°C. In this case the maximum temperature was attained in about 4 hours, and the test pieces, which were inserted into the furnace at the beginning, were still maintained at this maximum recrystallization temperature for 20 hours. The values of the grain numbers per unit area of the surface of the test pieces, averaged with a number of test pieces with equal amount of cold working respectively, are tabulated in Table II.

With the recrystallization temperature as high as at least 600°C or more, the grain growth is almost completed with prolonged heating, and the average grain number is almost independent from the maximum recrystallization temperature. The values tabulated in table II

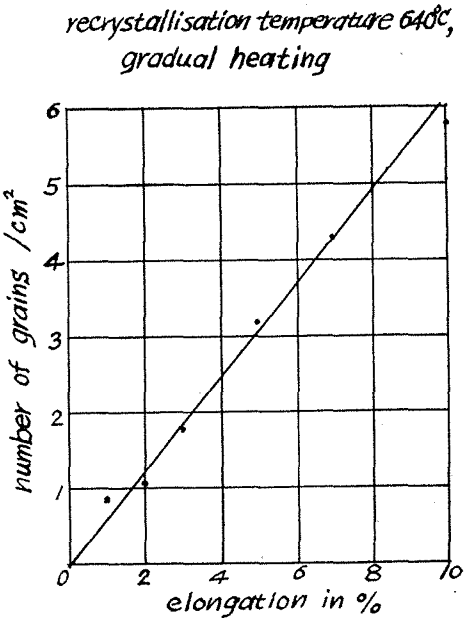
Table II

Maximum recrystallization temperature 640°C, (gradual heating)

Elongation in %	10	7	5	3	2	1
Number of grains /cm²	5.8	4.3	3.2	1.8	1.1	0.9

are shown graphically in Fig. 1, and a linear relation between the elongation and the number of grains per unit area is revealed clearly. The grain number observed in the present case is very much smaller and is of different order compared with the case of sudden heating. This point is very important, and we must be very cautious on this point in experimenting with the effect of temperature on the recrystallization phenomena; otherwise the results are liable to become entirely erroneous. For example if the thickness of the test piece is too large, it will be almost impossible to carry out the experiment with the effect of sudden heating, owing to the difficulty in applying a definite recrystallization temperature very quickly to the whole portion of the specimen.

Fig. 1



In the case of sudden heating to a recrystallization temperature, the number of grains per unit area increased with the recrystallization temperature for a given amount of cold working as stated before. But this is different from the recrystallization schema given by Czochralski[1] and

1. J. Czochralski: Moderne Metallkunde, 130, (1924).

other authors, where the grain size (not the grain number) is considered to become larger with increase of the recrystallization temperature. This discrepancy seems to be caused by the fact that the conditions under which they made their experiments rather belong to those of gradual heating, but not to those of sudden heating. In the present experiment the recrystallization temperature was higher than 500°C, and the crystal grains of aluminium obtained by recrystallization were rather large, so that the grain number could be counted by the naked eye. Consequently the recrystallization temperatures higher than 500°C for aluminium seems to be generally in germinative temperature range[1] in which exaggerated grain growth occurs, for the amount of cold working stated before, though the germinative temperature depends upon the amount of cold working. Thus it may be said, in the case of sudden heating to a recrystallization temperature, that the grain size diminishes with the increase of the recrystallization temperature, so far as the recrystallization temperature remain in the germinative temperature range.

Next the writers made a rough estimate of the time necessary for germination after the test piece had been subjected suddenly to a recrystallization temperature. For this purpose several test pieces were prepared for a given amount of elongation and a given recrystallization temperature. These test pieces were subjected separately to the same recrystallization temperature for different durations of time, and the state of the progress of germination was made perceptible to the naked eye by etching the surface of the test pieces with some reagent after they had been taken out of the furnace. The writers classified the recrystallized test pieces into three groups: 1) "no indication of germination," 2) "indication of germination" and 3) "complete germination." Among these three, 1) and 2) can be distinguished with adequate sharpness in recrystallization time, and the time necessary for the state 2) changes most regularly with the amount of cold working and the recrystallization temperature. On the contrary the time necessary for the state 3) changes very irregularly with tolerable fluctuation, which makes it almost impossible to set a sharp distinction between the states 2) and 3) with recrystallization time. Moreover X-ray examination indicates that already in the state 2) most of the metal crystals have grown into fairly large ones, being disclosed by the appearance of only a few Laue-spots of strong

1. Jefferies and Archer: The science of metals, 108 (1924).

intensity. Thus the time necessary to bring about the state of "indication of germination" was looked for roughly as the measure of the time which was necessary for germination to be almost completed, and it will be called "the germination time" hereafter. In the present experiment the germination time defined thus was measured in the unit of one minute; and the reciprocal of the germination time, which

Fig. 2

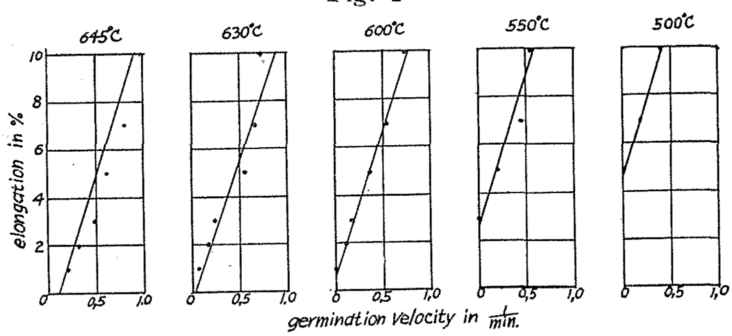
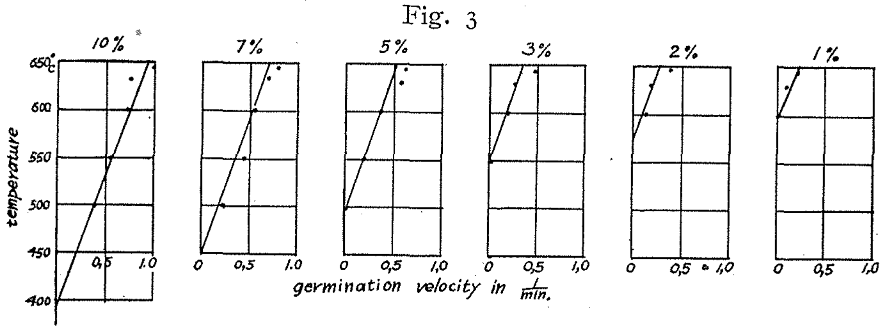


Fig. 3



may be called "the germination velocity," was considered in relation to the amount of cold working and to the germinative temperature. The relation between the elongation of the specimen and the germination velocity for various germinative temperatures is shown in Fig. 2, and the relation between the germinative temperature and the germination velocity for various amounts of cold working is represented in Fig. 3. From these figures it will be easily seen that the germination velocity increases almost linearly both with the amount of cold working and the germinative temperature. If we represent the germination velocity by v, the elongation by S, the melting point of aluminium by T_m (657°C), and the germinative temperature by T, then the above linear relation is found to be expressed very simply by the following empirical formula:

$$v=0.082\{S-0.042(T_m-T)+1.84\}\ldots\ldots\ldots\ldots(1).$$

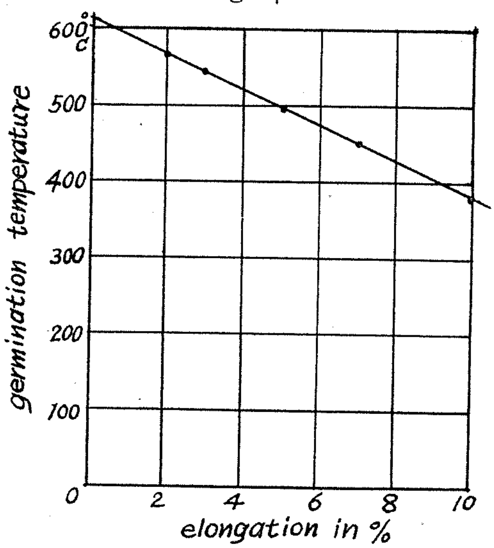
The relation given by this formula is represented by the straight lines drawn in Figs. 2 and 3, which fit fairly well with the observation.

It will be seen from the observation on germination velocity that the germination takes place at a temperature higher than a certain temperature, which may be called "the germination temperature," and that the germination temperature decreases with increase of the amount of cold working as in the case of the annealing temperature. If we represent by T_g the germination temperature, then the relation between T_g and the amount of cold working will be readily obtained by putting $v=0$, and $T=T_g$ in the above equation as follows,

$$T_m-T_g=\frac{S+1.84}{0.042}\ldots\ldots\ldots\ldots(2).$$

This relation is shown by the straight line in Fig. 4. Here it must be noted that the germination temperature, like the annealing temperature, is probably not a very definite one, and that more prolonged heating than in the present experiment may cause further development of germination. And consequently the straight line drawn in Fig. 4 ought to be understood to represent the higher limit of the rather indefinite germination temperature.

Fig. 4



From the standpoint of the theory stated before, the writers next attempted to formulate roughly the relation between the grain number, amount of cold working, and the germinative temperature to which the specimen was subjected for its germination. In that theory it was assumed that the crystal boundaries were easier to melt than in the interior of the crystal, and that the melting point of a crystal is lower with increased distortion, and that the recrystallization is nothing but the growth of the perfect crystal nuclei formed in the melt. It seems very probable that the grain growth to considerable size by recrystallisation does not take place directly by the consumption of the initial minute crystallites which have suffered some distortion by the previous cold working, but that it is performed by repeating several times (two times probably) the process of melting and crystallization, as is manifested by rather even or uniform grain growth of the metal crystallites at a rather lower recrystallization temperature. This seems to the writers to be due to the fact that at low temperature in the case of gradual heating or at early stage of recrystallization in the case of sudden heating, the crystals formed by recrystallization are not entirely free from strain; and thus they have still the predisposition to be recrystallized on further heating. When the temperature in the case of gradual heating becomes higher or when the stage of recrystallization advances further in the case of sudden heating, the specimen will get sufficiently soft, so that the new crystals formed then become almost entirely free from strain; and consequently they grow further and further on continued heating to exaggerated grain sizes by consuming the older crystals having more strains than themselves. The stage of germination seems to be understood to occur in such a manner.

Though the phenomena of recrystallization of metals are very complicated as stated above, it seems almost certain that the presence of distortion in the crystal lattice is its primary cause. When a small part of the crystal lattice is distorted the potential energy of the atoms in that part will become higher than the normal value in the same part, which is peculiar to the perfect crystal lattice. Next let us consider a small volume element in a distorted crystal lattice and the excess of the potential energy per unit volume at that part, in reference to the perfect undistorted lattice is ϕ. Then it seems not to be entirely absurd to assume that the number of localities $Nd\phi$ per unit area of the specimen which has a value of the excess potential energy ranging between ϕ and $\phi+d\phi$ is governed by a probability law, and that the localities having a larger value of ϕ increase in number with increase of the amount S of cold working.

This consideration may be roughly expressed by

$$Nd\phi = Ce^{-\frac{\Phi}{\alpha S}}d\phi \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (3).$$

Next assume that the lowering of the melting point at a locality in the crystal is proportional to the amount of the excess potential energy ϕ there. Thus if we represent by T the melting point at that locality, and by T_m the melting point of the perfect crystal, then equation (3) becomes

$$N \cdot dT = Ce^{-\frac{T_m - T}{aS}} dT \quad \text{.................................}(4),$$

where C and a are constant.

Now let us consider the case when the specimen is suddenly subjected to a recrystallization temperature, then the number of the crystal nuclei N' per unit area which is supposed to be formed at melted localities will be given by

$$N' = \int_0^T N \cdot dT = \int_0^T Ce^{-\frac{T_m - T}{aS}} dT = aSC\left(e^{-\frac{T_m - T}{aS}} - e^{-\frac{T_m}{aS}}\right).$$

If we consider the case when the recrystallization temperature is not very far from the normal melting point of the metal as is actually the case, then the second term in the bracket may be neglected in comparison with the first term, and the above equation becomes

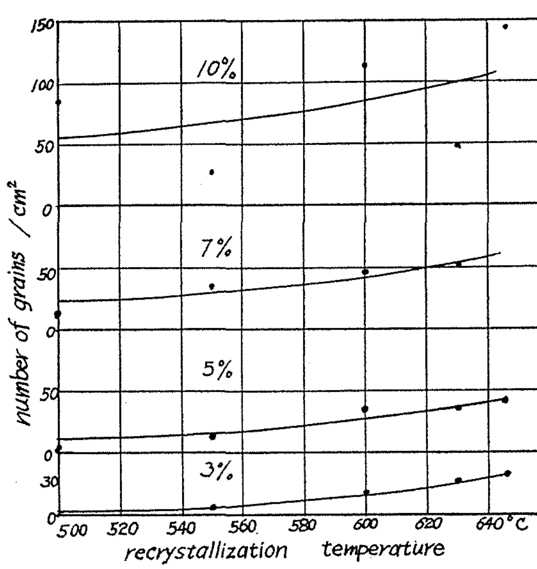
$$N' = aSCe^{-\frac{T_m - T}{aS}}.$$

Generally let us assume that C is not independent of the amount of the cold working, and represent aSC by a new constant K which is a function of S, then we obtain

$$N' = Ke^{-\frac{T_m - T}{aS}} \quad \text{.......................................}(5),$$

which represents the relation between the number of the nuclei per unit area of the surface of the specimen, the recrystallization temperature to which the specimen is subjected suddenly, and the amount of the cold working which is represented in the present case by the elongation expressed in % of the original length. As when the crystals are fully grown the number of the crystal nuclei becomes entirely equal to the grain number, the number of the crystal grains per unit area of the surface of the specimen is given by the above equation too.

Fig. 5

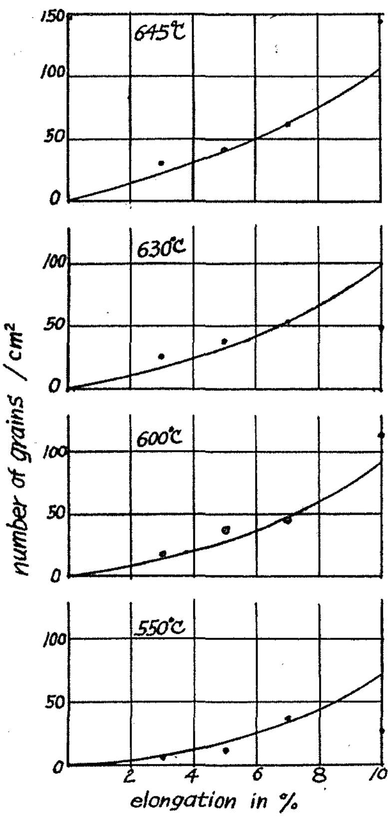


The constants a and K in the above equation were so chosen as to fit as near as possible to the grain number observed in the present experiment and tabulated in Table I for the case of sudden heating, and for their best values the writers obtained the following values by trial:

$$a=21.0, \quad K=21.0\times e^{0.18S}.$$

The values of N' calculated by putting these values to equation (5) are plotted in curves in Figs. 5 and 6. Though the agreement between the calculated and the observed values represented by dots is not satisfactory, the general tendency seems to be well expressed by the consideration stated before.

Fig. 6



It is already stated that the number of the crystal grains formed by recrystallization is very much different according as the specimen is subjected suddenly to a definite recrystallization temperature or the recrystallization temperature is raised gradually from the room temperature to a final value, even when this final value is the same as the recrystallization temperature in the case of sudden heating. This fact indicates that the grain number depends more upon the heating at lower temperatures than the final maximum temperature in the case of gradual heating. To make clear this point the writers carried out another experiment. Test pieces of commercial aluminium plate having the same size as stated before were preliminary annealed for 24 hours at the temperature of 320°C, then they were elongated 7% of the original length. After these test pieces were subjected suddenly to the first lower recrystallization temperature separately for about 5 hours, also their temperature was raised gradually to the second higher recrystallization temperature of 645°C for about 20 hours separately. The number of the grains thus formed per square centimetre of the surface of the specimen, corresponding to various first recrystallization temperatures are tabulated in Table III.

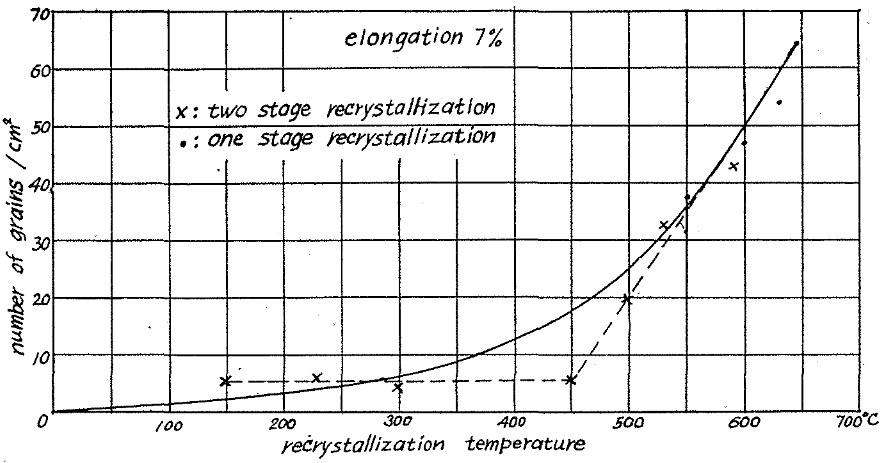
Table III

First recrystallization temperature, °C	590°	530°	500°	450°	300°	230°	150°
Number of grains/cm²	43	33	20	6.0	4.6	5.8	5.5

Elongation 7%, second recrystallization temperature 645°C.

In order to compare the results obtained now with two stage recrystallization to those obtained before with one stage recrystallization, the results obtained in both cases are represented together in Fig. 7, by regarding the first lower recrystallization temperature in the case of two stage recrystallization as the same as the recrystallization temperature in the case of one stage recrystallization. Nice amalgamation of the two results, as is seen in the figure, indicates clearly that the number of the crystal grains in the case of two stage heating, when

Fig. 7



they are pretty well developed after being exposed to the second higher recrystallization temperature belonging to the germinative temperature rang, is primarily determined by the first lower recrystallization temperature. This seems to indicate that the healthy perfect crystal nuclei which have the ability to grow further and further in the germinative temperature range, or the localities where such nuclei are to be formed are already seeded in the first stage of lower recrystallization temperature; and that the germination is nothing but the growth of such healthy crystal nuclei. In Fig. 7 the curve drawn in full line is that calculated by equation (5), and the broken straight lines are so drawn that they may fit best to the observation points. These two broken straight lines intersect at about 450°C, which is nothing but the germination temperature for the elongation of 7% as will be seen from Fig. 4. Thus it may be said that in the germinative temperature range, seeding and germination of the healthy crystal nuclei occur simultaneously in the case of sudden heating, and that in the recrystallization temperature range lower than the germination temperature seeding only without germination takes place. Though the number of the crystals recrystallized in such a lower recrystallization temperature range is immense, the number of the seeds formed in such a stage is very small only of the order of about 5 per square centimetre on the surface of the aluminium plate in the present case. This number coincides roughly with that obtained by extrapolating the theoretical curve in Fig. 7, and also with that obtained in the case of gradual heating for 7% elongation, which was about 4 as is given in Table II. This seems to indicate that in the case of gradual heating up to the germinative temperature range, only a few seeds formed among an immense number of the crystals recrystallized in the early stage of lower temperature grow to very large crystals in the later germinative temperature range. The question of why a much larger number of grains are formed in the case of sudden heating in the germinative temperature range than in the case of gradual heating seems to be understood in such a manner.

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