A Study of the Rearrangement of the Crystals of a Metal by Its Recrystallization

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Abstract

By means of an X-ray examination of thin plates of aluminium, it was observed that the germination process, in the case of sudden heating to the germinative temperature range, took place in two stages with a cold worked aluminium plate: primary grain growth to a moderate size starting from a very minute one, and then the secondary grain growth to exaggerated size starting from the moderate one. As to the orientation of the crystal formed by recrystallizing elongated thin aluminium plate consisting of its single crystal with gradual heating, it was found that when the elongation was less than about 2 percent the new crystal formed by recrystallization retained one of the orientation before recrystallization which had been occupied by one of the micro-crystals formed by crushing the single crystal by the process of stretching; and that when the elongation was greater than about 2 or 3 percent the orientation of the new crystal formed by recrystallization was independent from those of the micro-crystals before recrystallization.

In a paper[1] recently published the writers stated that the grain growth of metal crystals to a considerable size by recrystallization in the germinative temperature range did not take place directly by consuming the initial minute crystallites which had suffered some distortion by a previous cold working, but that it was produced by repeating several times (two times probably) the process of melting and crystallization, as was manifested by rather even or uniform grain growth of the metal crystallites at a rather lower recrystallization temperature. This point being very important in understanding the process of recrystallization of metals, the present experiment to elucidate it was carried out with thin commercial aluminium plate. The size of all the test pieces was 10 cm × 1.5 cm × 1 mm. These test pieces were first annealed for 48 hours at the temperature of 320°C, and then were stretched in various degrees of elongation ranging from 1 to 5 percent. Each test piece was then put suddenly, and for various durations of time, into a furnace always kept at the temperature of 630°C. This temperature is certainly within the germinative temperature range for

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[1] U. Yoshida, S. Nagata and Ch. Mitsuki: These Memoirs, **19**, 169 (1936).

the test pieces stated above. As it was necessary in the present case to inspect with X-rays the manner of the grain growth in the intermediate stage of recrystallization, each test piece was subjected suddenly and successively to the same recrystallization temperature for various durations of recrystallizing time ranging from twenty or thirty seconds to several minutes by inserting into and taking out of the furnace alternately. An X-ray diffraction photograph was taken at every stage of manipulation.

Table I

State of the test piece	Manipulation of the test piece	State of the test piece	Manipulation of the test piece
a	commercial Al plate	*g*	*f* annealed at 630°C, 4 minutes
b	*a* annealed at 320°C, 24 hours	*h*	*g* annealed at 630°C, 2 minutes
c	*b* elongated 1.3%	*i*	*h* annealed at 630°C, 3 minutes
d	*c* annealed at 630°C, 20 seconds	*j*	*i* annealed at 630°C, 5 minutes
e	*d* annealed at 630°C, 1 minute	*k*	*j* annealed at 630°C, 5 minutes
f	*e* annealed at 630°C, 2 minutes		

An example of such manipulation of a test piece elongated by 1.3 percent is shown by Table I, and some of the X-ray photographs taken at the different stages of manipulation are reproduced in Fig. 1, Plate I. Any one of the photographs represented by *a*, *c*, *h* and *k* corresponds respectively to the stage of the test piece which is denoted by the same letter in Table I. Thus Fig. 1_a is obtained from the original commercial aluminium plate, Fig. 1_c from the test piece elongated 1.3 percent after the annealing at 320°C for 24 hours, Fig. 1_h from that subjected five times successively to the higher recrystallization temperature of 630°C, and Fig. 1_k from that subjected more three times successively to the same recrystallization temperature. In this last *k* state many minute Laue-spots, which were visible in earlier stages of recrystallization, were almost entirely displaced by a few stronger ones. This indicates that the germination is almost completed in this state, and any change in the distribution and the intensity of the Laue-spots was not detected on more prolonged subjection to the same recrystallization temperature. Fig. 1_h corresponds to an intermediate stage of the germination; and this photograph shows us, in addition to a few stronger Laue-spots which are the same as those seen in Fig. 1_k, the presence of a large number of minute and distinct Laue-spots whose appearance is clearly different from those in Fig. 1_c.

The diffused appearance of the Laue-spots, and consequently a rather continuous impression in Fig. 1_c, is of course due to the presence of an immense number of very tiny crystallites scattered rather evenly to some extent in their orientations. These very tiny crystallites grow up first into larger ones, as is observed by the presence of a large number of minute and distinct Laue-spots in Fig. 1_h; and then the crystallites thus grown develop to one or a few adult crystals in the process of germination, as is shown by the presence of a few strong Laue-spots in Fig. 1_k. From these facts it does not seem absurd to say that the growth of the crystals in the germination of a cold worked metal takes place roughly in two stages: first, from very tiny crystallites to those of intermediate size, and then to the adult size from the intermediate one. Here it must be noted that, in taking the Laue-photographs at various stages of manipulation of a test piece, special care was always taken to illuminate the same portion of the specimen with the pencil of X-rays. The photographs reproduced in Fig. 2, Plate I represent another example of the manner of crystal growth in the process of germination of a commercial aluminium plate, which is the same as in the case of Fig. 1. These photographs were taken by the long slit method so as to be capable of revealing a larger portion at once. Fig. 2_a was taken with a specimen obtained by annealing a commercial plate of aluminium at 320°C for 24 hours, by elongating it 3 percent and then by subjecting it to the recrystallization temperature of 620°C for 1.5 minute. With this short duration of recrystallizing time no indication of recrystallization could be detected. Fig. 2_b was obtained when the same test piece was heated suddenly for 2.5 minutes more at the same recrystallization temperature. It corresponds to an intermediate stage of germination. Fig. 2_c was obtained when the same test piece was heated still more suddenly for 5 minutes at the same recrystallization temperature. This corresponds to the stage of completed germination. These photographs seem to reveal the same aspect of the crystal growth in the germination process as in the case of Fig. 1, Plate I. Moreover, a close inspection of Fig. 2_b and Fig. 2_c shows us the manner of the crystal growth minutely in an interesting manner.

Next, the writers prepared many single crystal plates of aluminium by the ordinary stress-annealing method, and stretched them separately in various degrees of elongation ranging from 0.9 to 9 percent of the original length. These plates were then recrystallized at a temperature of about 610°C for various durations of time ranging from several to about 20 hours, the temperature being raised gradually from the room temperature. The orientation of the aluminium crystals thus formed by recrystallization was examined by taking Laue-photographs of the specimens before and after the recrystallization respectively. A mere comparison of the two Laue-photographs taken before and after the recrystallization of the same portion of the specimen, will show clearly whether the orientation of the new crystal formed by recrystallization changed from or recovered that of the crystal before recrystallization. Three examples of such Laue-photographs are reproduced in Figs. 3, 4 and 5 in Plate II. In the case of Fig. 3, where the elongation was 2%, the photographs were taken by the long slit method. In this plate, Fig. 3_a corresponds to the original single crystal plate, Fig. 3_c to the recrystallized one and Fig. 3_b to the elongated one before recrystallization. The scattering of the orientations of the crystallites formed by crushing the original single crystal by the process of elongation is sufficiently perceptible in the diffuseness of the Laue-lines in Fig. 3_b. Such diffuse Laue-lines became perfectly sharp with recrystallization, as is seen in Fig. 3_c. Their position in this figure coincides perfectly with those in Fig. 3_a, showing that the orientation of the new crystal formed by recrystallization recovered that of the original crystal before recrystallization. The photographs reproduced in Figs. 4 and 5 in Plate II were taken by the ordinary Laue-method. Figs. 4_a and 5_a correspond to the elongated single crystal plates before recrystallization, and Figs. 4_b and 5_b correspond to the recrystallized plates respectively. In the case of Fig. 4 the photographs show that the orientation of the new crystal formed by recrystallization recovered that occupied by some one of the crystallites in the elongated test piece before recrystallization, as in the case of Fig. 3. Contrary to such examples the photographs in Fig. 5 show that the new crystal formed by recrystallization took a new orientation which was entirely different from those of the crystallites in the elongated plate before recrystallization. The writers carried out the same experiment with many single crystal plates of aluminium which were elongated differently, and the results obtained are tabulated in Table II. As is seen in the table, the orientation of the new crystal formed by recrystallization recovered, with only one exception, that of some one of the crystallites before recrystallization when the elongation was less than about 2%, while it changed always by recrystallization when the elongation

Table II

Elongation in %	Orientation of the recrystallized crystal	Elongation in %	Orientation of the recrystallized crystal
9.2	changed	2.7	changed
9.1	changed	2.6	recovered
7.4	2 crystals, both changed	2.5	2 crystals, one recovered, the other changed
6.1	changed	2.0	recovered
5.2	many crystals	1.6	recovered
5.2	changed	1.5	recovered
3.7	changed	1.5	recovered
3.6	2 crystals, both changed	1.4	recovered
3.2	2 crystals, both changed	1.3	recovered
3.1	recovered	1.1	2 crystals, one recovered, the other changed
2.9	changed	0.9	recovered

was greater than about 3.2%; and with elongation between these two limits, the results were mixed. Before this experiment a similar one with aluminium was performed by K. Tanaka[1] who elongated the single crystals within the range of 4–20 percent, and found that the new crystal formed by recrystallization took a new orientation which was rather independent from those of the crystallites before recrystallization. Thus his result agrees with the present when the elongation is greater than about 3 or 4 percent.

In a theory of the recrystallization of metals[2] proposed by one of the writers and Koyanagi, it was assumed that distorted metal crystals were easier to melt, and that the boundaries of metal crystals were more easily melted than their interior; and the independence of the orientation of the new crystals from those of the crystallites before recrystallization was considered to be due to the spontaneous formation of new crystal nuclei in the melt at the crystal boundaries. Though the results obtained in the present experiment as stated above seem to contradict to some extent this consideration, actually they do not. The results may be made clear by a detailed explanation of the theory. When the elongation of the single crystal plate is little, we can expect the persistence of a few undistorted small crystal fragments among an immense number of distorted fragments. If such is the case, it seems natural to suppose that some of the crystal fragments lying just in the immediate neighbourhood of such undistorted frag-

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[1] K. Tanaka: These Memoirs, **11**, 229 (1928); and **13**, 117 (1930).

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

ments are pretty severely distorted. If the temperature of such specimen is gradually raised to its recrystallization temperature, the severely distorted crystal fragments will melt and immediately adhere to the neighbouring undistorted fragments, resulting in the growth of the undistorted crystal fragment without the formation of any new crystal nucleus in the melt. Consequently the undistorted crystal fragment itself becomes the recrystallization nucleus in this case and the process of recrystallization proceeds from this nucleus. The recovery of crystal orientation by recrystallization with a little elongation is understood to take place in such a manner. When the elongation is much greater the persistence of such undistorted crystal fragments is not conceivable, and the recrystallization nucleus ought to be formed spontaneously in the melt at the crystal boundaries in the process of recrystallization; and thus the orientation of the new crystals becomes independent of those of the crystallites before recrystallization.

A similar experiment on the orientation of the crystals formed by recrystallizing elongated aluminium single crystal plates was also performed by suddenly subjecting the specimens to the recrystallization temperature of 610°C. But so far as the present experiment is concerned, the writers could not observe that the recrystallized crystals recovered the orientation of the old crystallites even with the elongations less than 2 percent. Moreover the number of the crystals formed by recrystallization was generally larger than in the case of gradual heating, as was expected by the former experiment[1] on the effect of sudden heating upon the grain size. In the case of sudden heating, the localities in the specimen, where the distorted crystallites melt and turn out to the recrystallization nuclei, become immense, as was stated in the former paper; and the recrystallization nuclei are formed spontaneously at the localities other than the immediate neighbourhood of undistorted crystal fragments. This seems to be the reason for the difficulty of the recrystallized crystals to recover orientation when the specimen is heated suddenly to its germination temperature. In the paper[2] cited a recrystallization experiment was described in which a cold worked aluminium plate was first subjected suddenly to a lower recrystallization temperature for some hours, after which the temperature was raised gradually up to a second higher recrystallization temperature belonging to the germinative temperature range. From

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[1] U. Yoshida, S. Nagata and Ch. Mitsuki: These Memoirs, **19**, 169, (1936).

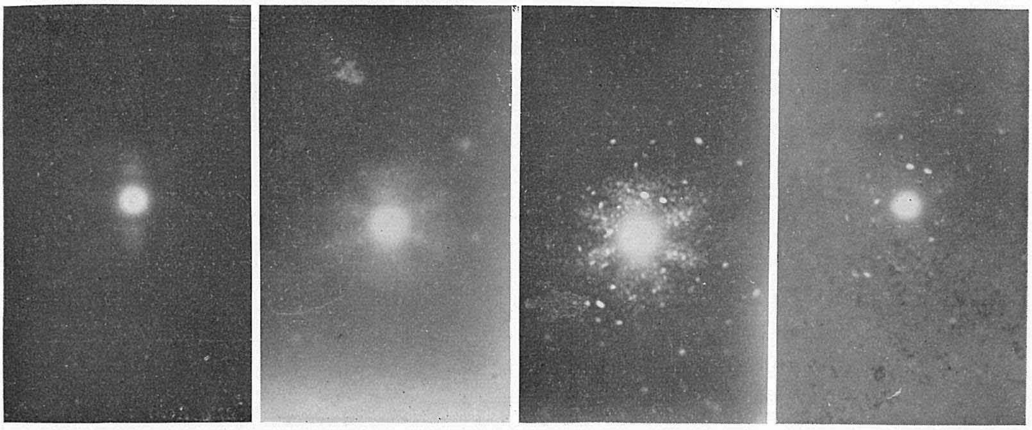
[2] ibid.

the number of the crystals thus formed per unit area on the surface of the specimen, it was considered that the healthy perfect crystal nuclei which had the ability to grow further and further in the germinative temperature range, or the localities where such nuclei were to be formed were already seeded in the first stage of lower recrystallization temperature; and that the germination was nothing but the growth of such healthy crystal nuclei. This rather ambiguous idea regarding the seeding of the crystal nuclei seems much clearer now in the present experiment on the recovery of the orientation of the recrystallized crystals, and it seems more reasonable to consider that the seeding is nothing but the formation of some undistorted crystal nuclei in the lower recrystallization temperature, which have the ability to grow further and further in the germinative temperature range.

In conclusion, the writers' sincere thanks are due to the Hattori Hoko Kai for their grant given to one of the writers (U. Y.), with the aid of which this research was accomplished.

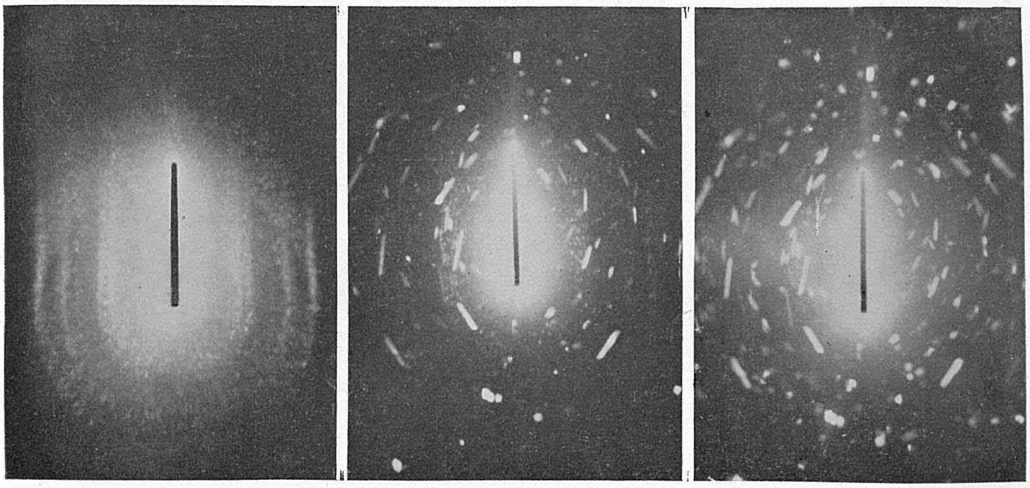
Plate I

Fig. 1



a *c* *h* *k*

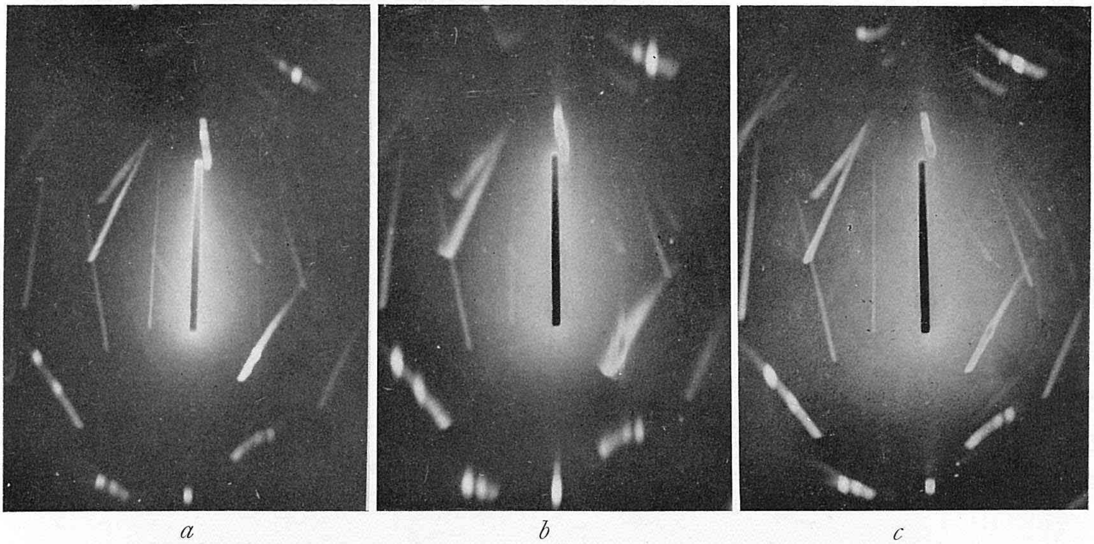
Fig. 2



a *b* *c*

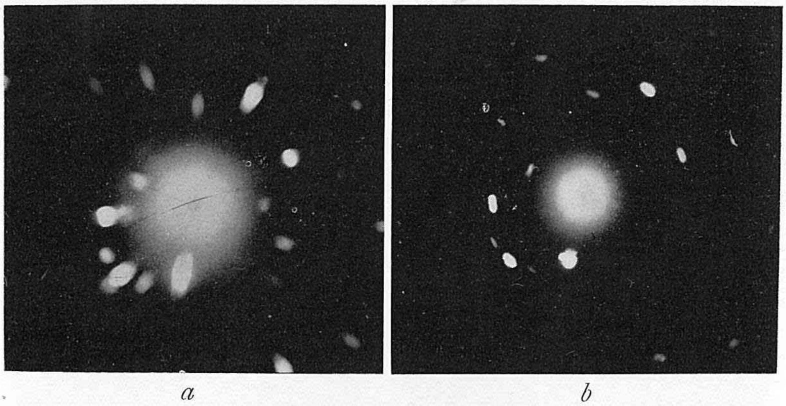
Plate II

Fig. 3 (Elongation 2%)



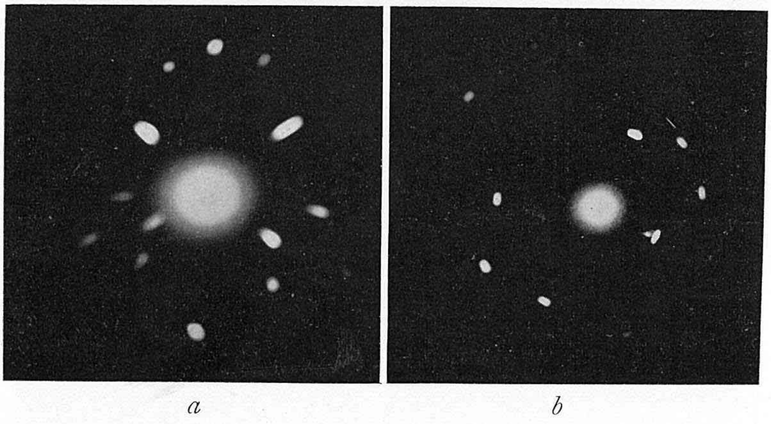
a *b* *c*

Fig. 4 (Elongation 2%)



a *b*

Fig. 5 (Elongation 2.7%)



a *b*