# Research on the Structure and Low-Temperature Annealing Effect in Cold-Rolled $\alpha$-Brass. I
## Structure and Change in Hardness*

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**Synopsis**

The change in hardness by low-temperature annealing of cold-rolled 70/30 brasses having different ready-to-finish grain sizes (fine and coarse) and cold-rolled by different reductions (10~90 per cent) was measured. The specimens were annealed at 150, 250, 350 and 450° for 5 min~24 hrs. The hardness change by annealing was considered to be related to the structures as ready-to-finish annealed and as cold-rolled. It appears that the hardness does not vary simply with rolling reduction or grain size, but is more complicatedly influenced by the development degree of the rolling structure or by the condition of specimens such as the composition.

## I. Introduction

The primary slip-lines, which developed in individual grain of 70/30 brass by slight cold-rolling, gradually changes into complicated forms, the secondary slip-lines, with increasing rolling reduction, which, however, reaches the limit at about 50 per cent in reduction. The more the reduction increases, the more remarkable becomes the surface marking of deformation which is perpendicular to the rolling direction on the sheet surface and tilted by 30~40° to the surface in the longitudinal section. In addition, it is said that the grain size brings out a little bit of different result in the development of rolling structure. As to the grain size, though a considerable attention has been paid to an annealed material from the technical point of the grain size control, yet there are few researches treating of the subject from the cold-worked structure. Especially, there are very few systematic investigations on an anneal-hardening of brass from this standpoint.

The present study, therefore, concerned with the hardness change due to annealing, both in the anneal-hardening and in the recrystallization range, of cold-rolled 70/30 brasses with different grain sizes and reductions.

## II. Experimental procedures

### 1. Specimen

All specimens were commercially pure 70/30 brass and finally rolled to 0.5 mm in thickness with a working procedure shown in Table 1, that is, the specimen

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Table 1. Working procedures of specimen.

| | | |
|---|---|---|
| A | 6mm→500°×3hr→2.20mm→500°×3hr→1.10mm→500°×3hr→0.55mm→ | {500°×3hr→0.5mm<br>750°×3hr→0.5mm |
| B | 6mm→500°×3hr→2.84mm→500°×3hr→1.42mm→500°×3hr→0.71mm→ | {500°×3hr→0.5mm<br>750°×3hr→0.5mm |
| C | 6mm→500°×3hr→4.00mm→500°×3hr→2.00mm→500°×3hr→1.00mm→ | {500°×3hr→0.5mm<br>750°×3hr→0.5mm |
| D | 6mm→500°×3hr→3.40mm→500°×3hr→1.70mm→ | {500°×3hr→0.5mm<br>750°×3hr→0.5mm |
| E | 6mm→ | {500°×3hr→0.5mm<br>750°×3hr→0.5mm |

group of each reduction was ready-to-finish annealed at 500° and 750°, resulting in a fine (0.02~0.03 mm) and a coarse (0.15~0.20 mm) grained one, respectively, and then was handed to the final cold-rolling of various reductions. The cold-rolling was carried out all in straight rolling. The chemical composition of specimens is shown in Table 2. For comparison, electrolytic copper melted in vacuum was

Table 2. Chemical composition of specimens.

| Specimen | Composition, wt. % | | | | |
|---|---|---|---|---|---|
| | Zn | Fe | Pb | P | Cu |
| H | 31.36 | 0.003 | 0.007 | 0.0002 | R |
| W | 31.21 | 0.009 | 0.020 | 0.0005 | R |

also used after the final rolling of 50 and 80 per cent reduction (the oxygen content <0.002 per cent).

2. Experimental method

(i) Annealing conditions

From the above-mentioned rolled plate was cut off the specimen of 20×30(mm$^2$) in dimension. Each specimen was heated in a small electric muffle furnace kept at 150, 250, 350 and 450° and after keeping for a certain time it was air-cooled. Brass specimen was packed with brass foil, and copper specimen with copper foil, to prevent them from dezincification or oxidation as much as possible.

(ii) Measurement of hardness

The surface of the specimen was sufficiently polished to remove oxidized and dezincificated layers produced by heat-treatment. The hardness was measured with Vickers micro-hardness tester under 500 g load. To investigate furthermore the directionality, the directions of two diagonal lines of the Vickers indentations were arranged respectively parallel and normal to the rolling direction, and each mean value was taken. The average was taken of the values in the range within ±5 per cent respectively on 10 measured points. The measured values of material of low working reduction had a tendency to be comparatively sporadic. In the following figures, the mark ● refers to the hardness parallel to the rolling direction, whereas the mark + to that normal to the rolling direction.

## III. Experimental results and considerations

Of the four kinds of experiment the heat-treatment at 150° showed anneal-hardening without any recrystallization, and only in the specimens highly reduced recrystallization happened to take place at 250° after a certain time, and at 350° and 450° the specimens except those of low reduction showed the recrystallization-softening at the early stage of annealing. The details will be stated in the following.

1. Annealing effect at 150°

Fig. 1 shows an example of the hardness change in brass heated at 150° for

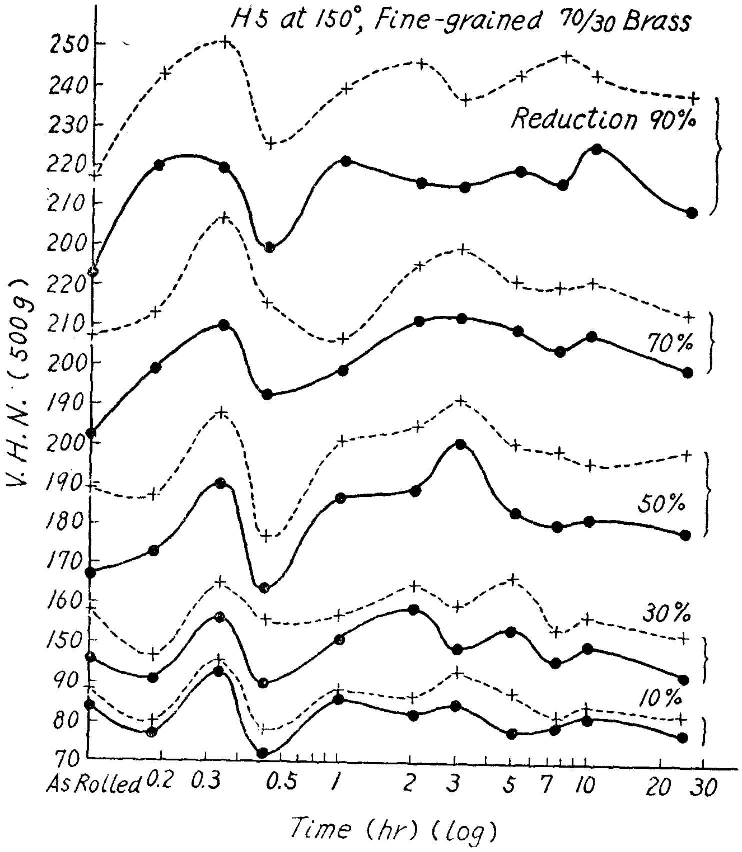


Fig. 1. Change in hardness at 150° of fine-grained, cold-rolled 70/30 brass, ● : Parallel to rolling direction, + : Transverse to rolling direction.

10 min ~24 hrs after the final rolling of 10~90 per cent reduction. At the early stage of annealing of about 20 min the primary hardening is seen, but when the working reduction increases, the maximum point of hardness tends to shift slightly to the short-time side. After passing through this stage the hardness changes goes through the softening process, and then transfers to the secondary hardening stage, until it reaches the highest value in 1~3 hr. The hardness in fine-grained specimen decreases a little when annealed for a long time, while the coarse grained specimen, after the secondary hardening, becomes once softened in 3~5 hr and then is hardened again as if a third hardening-stage exists. Such a change in

hardness was seen to shift a little to the long-time side in the case of a somewhat impure specimen (W). (The figure is omitted.)

Now, Fig. 2(a) shows the difference between the hardness in rolled state and

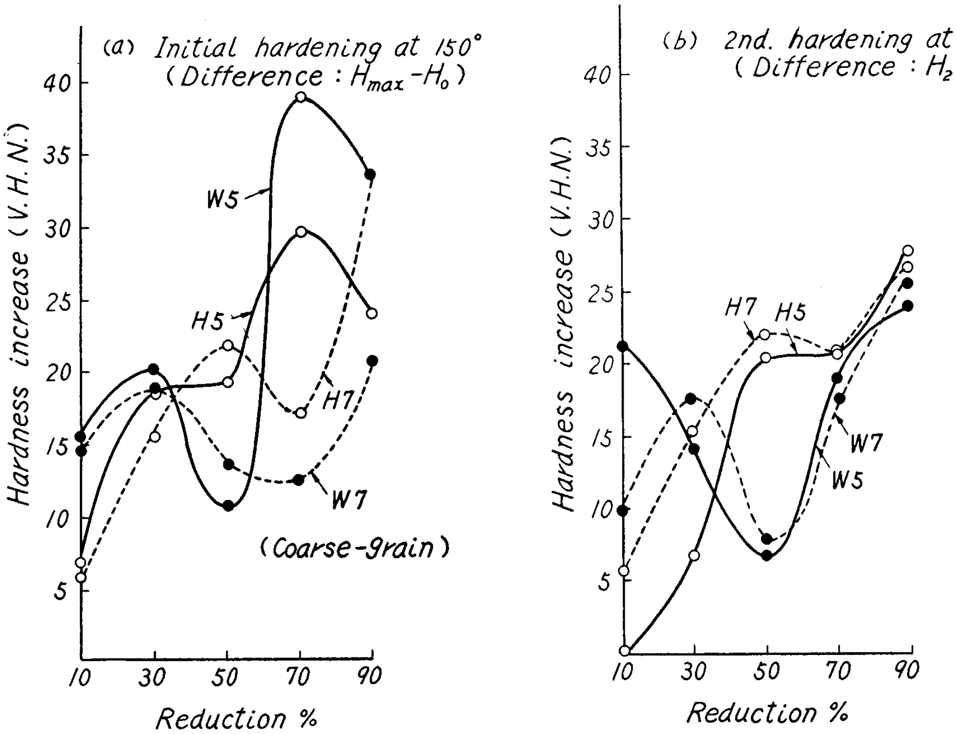


Fig. 2. Primary and secondary hardening at 150° of cold-rolled 70/30 brass sheets.

that maximum at the primary stage, the hardness being measured in the direction normal to the rolling direction. As seen in the figure, the amount of the primary hardening does not increase continuously with the increase of the working reduction. In the case of the fine-grained specimen (the full line in the figure), the hardening once increases with the increase of the working reduction in the range of low reduction and reaches the maximum at 30 per cent. When the working reduction becomes 50 per cent, the hardening decreases somewhat, forming a valley, and thereafter it increases rapidly though decreases a little at 90 per cent reduction compared with the case of 70 per cent. On the other hand, in the case of the coarse-grained specimen (the dotted line in the figure), the above-mentioned tendency seems to shift towards the high reduction side, and the increase still continues even at 90 per cent reduction. If this phenomenon is further examined in connection with rolling structures, an interesting relationship will be seen between them. Though the details of the structure will be reported later, in fine-grained specimens the 50 per cent reduction is the limit at which the slip deformation in individual grain is almost saturated, and 90 per cent in reduction is the stage at which the deformation due to a remarkable development of strain markings regardless of the grain boundaries reaches almost its limit. This change in deformation structure with increasing reduction tends to be retarted a little for the coarse-grained specimens. It can be said, therefore, that the development degree of the deformation structure, that is, the state and the uniformity of the strain distribution have a considerable influence on the primary hardening.

Fig. 2(b) shows a similar relationship in the secondary hardening process. In this case, a causal relation due to grain sizes or the development degree of deformation structures could not be perceived as in the case of the primary hardening, but the material conditions such as alloy's compositions or impurities were thought to be decisive factors, that is, the change in hardness would show a similar tendency if the specimens (H or W) were the same. Therefore, the primary hardening has a character of redistributing the stresses attendant upon the heat-treatment, while the secondary hardening is considered to be a reactional process affected by the behavior of solute atoms.

2. Annealing effect at 250°

When the specimens of different reductions were heated at 250° for 24 hours, no recrystallization occurred at the working reduction below 20 per cent, but above 50 per cent the recrystallization process began after a certain time. The tendency of the hardness change was quite different from that in the above case of 150°, and the hardening process with several stages was not observed at examined time intervals. The specimens rolled more than 50 per cent in reduction showed anneal-hardening before recrystallization, and the annealing time necessary for reaching the maximum hardness shifted to the short-time side in proportion

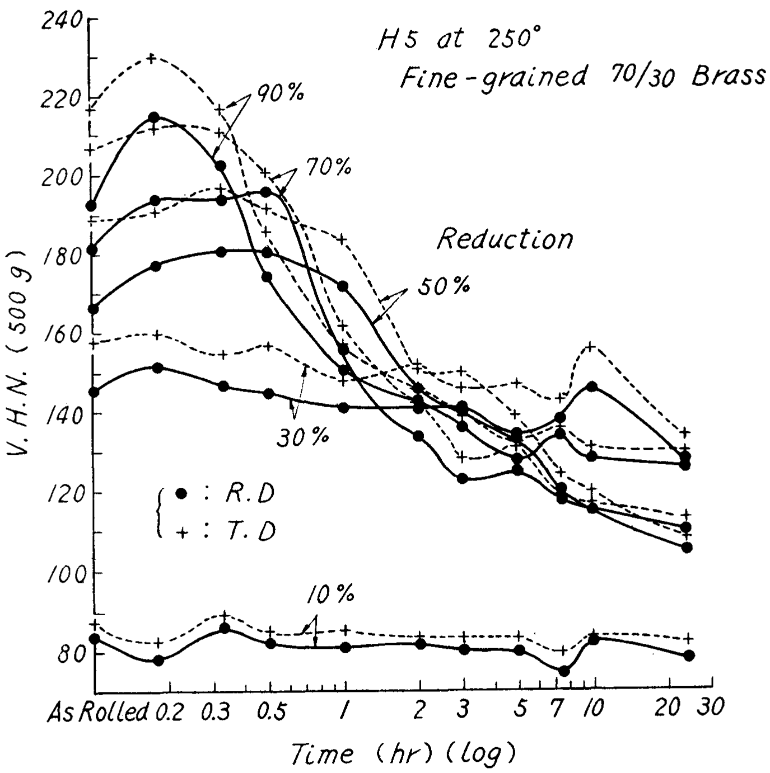


Fig. 3. Change in haıdness at 250° of fine-grained, cold-rolled 70/30 brass.

to the increase of the working reduction. It was also the case with the beginning time of recrystallization, but it tended to delay in the coarse-grained specimen. The change in hardness of fine-grained materials at 250° is shown in Fig. 3. The amount of anneal-hardening at 250° is much smaller than that at 150°. The hardening of each specimen is shown by a diagram in Fig. 4 for comparison's sake. Apparently different from the case at 150°, the change at 250° showed random tendencies in low-reduction range. Though the effect of the grain sizes was not perceived, the hardening gradually increased as the reduction goes up enough. The details will be stated later, but in the annealing at 250° the progress of a kind of polygonization was observed. The relation between this and the anneal-hardening, however, is not yet clear. In coarse-grained specimens the softening process was more retarded after the recrystallization took place.

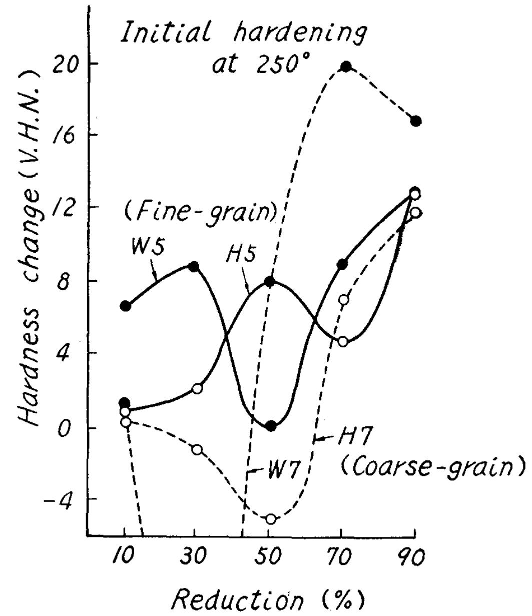


Fig. 4. Initial hardening at 250° of cold-rolled 70/30 brass sheets.

3. Annealing effect at 350°

In the annealing at 350° the specimens except those of 10 per cent in

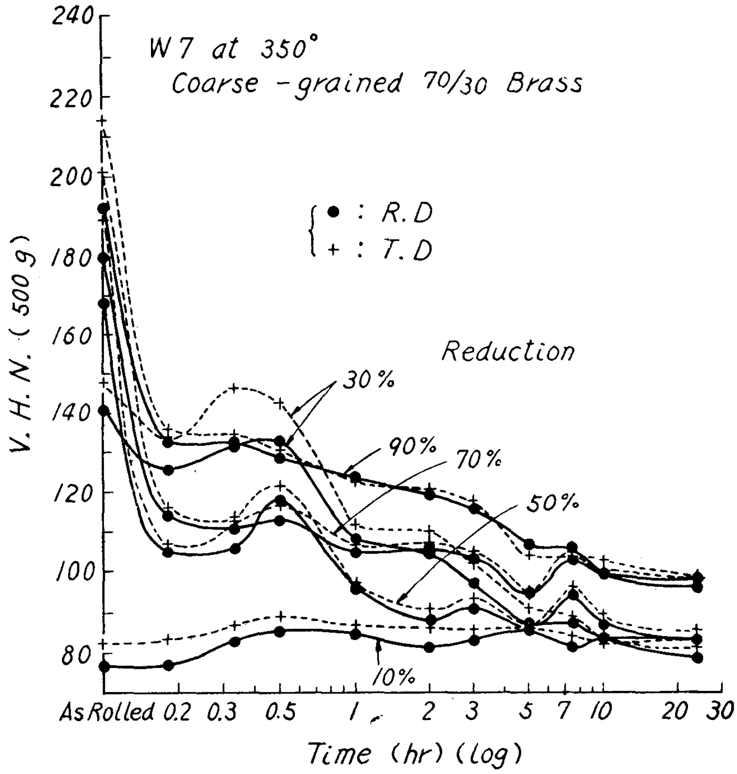


Fig. 5. Change in hardness at 350° of coarse-grained, cold-rolled 70/30 brass.

reduction began to recrystallize at an initial stage of heating, and the hardness changed through a softening process, that is, the hardness change is not continuous showing, at first, rapid softening, then slight hardening and finally again softening. The hardness change in coarse-grained materials is shown in Fig. 5. From the figure it will be seen that the lower the working reduction is, the more remarkable is the discontinuity of the above-mentioned hardness change. As to the initial stage of anneal-softening, the absolute values of hardness of each specimen are compared with one another as shown in Fig. 6(a). Apart from the specimens of

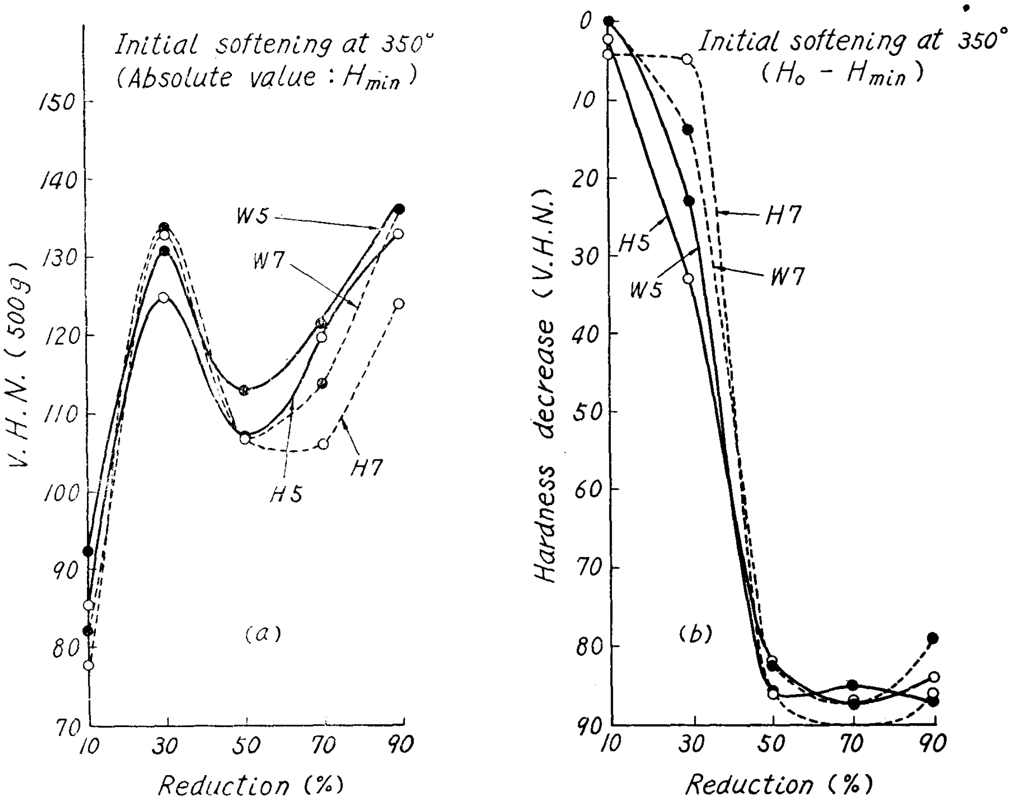


Fig. 6. Initial softening at 350° of cold-rolled 70/30 brass.

10 per cent in reduction, the absolute value of the hardess at the initial stage of softening shows the maximum at 30 per cent in reduction, and becomes mimimum at 50 per cent, and thereafter rises again with the increase of the working reduction. These differences in hardness between the specimens as rolled and initially softened, namely, the softenings are shown in Fig. 6(b). It is quite natural from the viewpoint of the working structure that the softening at the initial stage should delay in the material of 30 per cent reduction, but in the reduction over 50 per cent the softening remains almost unchanged, keeping the hardness difference due to the work-hardening. It is not clear that this phenomenon is explained whether by the fact that the strong tendency of hardening after softening increases with increasing reduction, so that the tendencies of softening and hardening offset each other, or by the fact that at the reduction above a certain amount the velocity of recrystallization at 350° is almost unchangeable. The ready-to-finish grain size has no remarkable influence upon it.

As for the hardening stage following the softening, the absolute values of the hardness showed the maximum at 30 per cent in reduction, as shown in Fig. 7(a), and in proportion to the increase in reduction, the fine-grained specimen showed

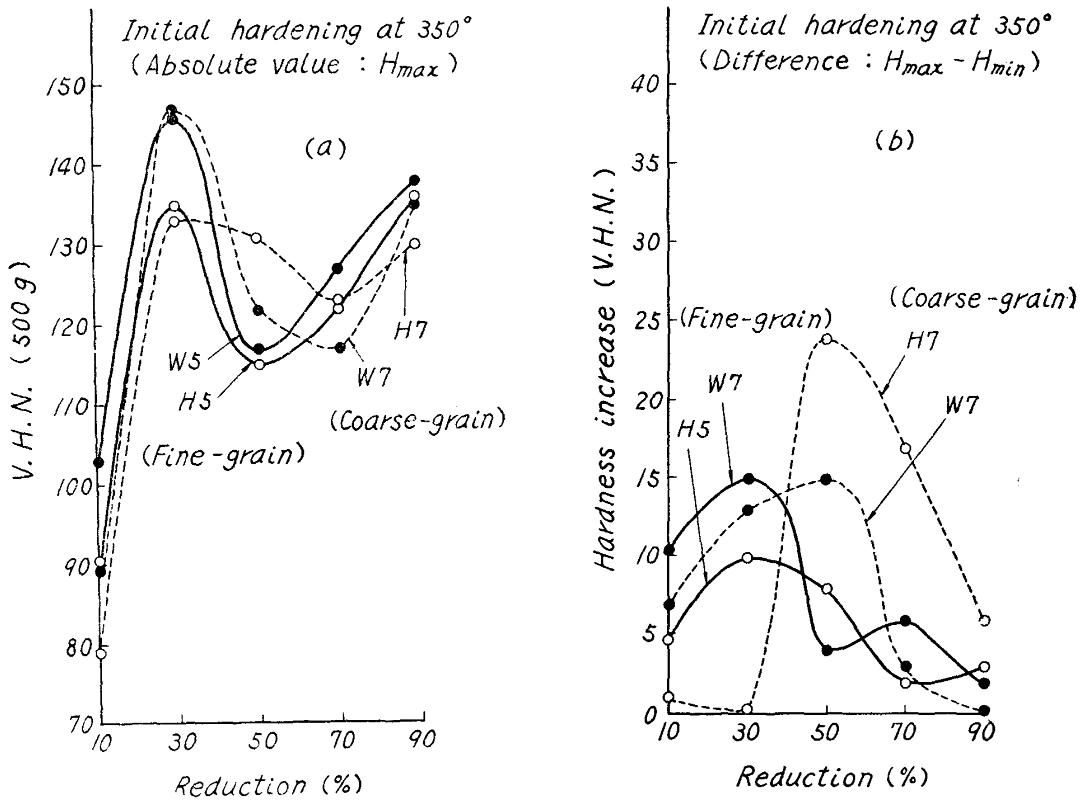


Fig. 7. Initial hardening at 350° of cold-rolled 70/30 brass.

the minimum at 50 per cent, while the coarse-grained one at 70 per cent, and further, the more highly the material is reduced, the more the value increases. When the difference between the hardness in this stage and that in the initial softening is taken as the measure of hardening, the greatest value is shown at 30 per cent in the fine-grained material, and at 50 per cent in the coarse-grained material, as is obvious from Fig. 7(b). The change in this stage is also influenced like this by the ready-to-finish grain sizes. That in the highly reduced material the hardening decreases in proportion to the increase of the working reduction might be due to the offset of the hardening tendency to the softening one in the same way as the case of the above-mentioned initial softening stage. Apart from the material of 10 per cent in reduction, after this hardening stage, the hardness changed through the softening process, and the hardness difference between specimens gradually decreased.

4. Annealing effect at 450°

At the annealing-temperature of 450° the hardness in each specimen changed through the softening process from the beginning of heat-treatment, but in coarse-grained materials the softening progressed more slowly. An example for this is shown in Fig. 8. Though the material of 10 per cent in reduction also showed a

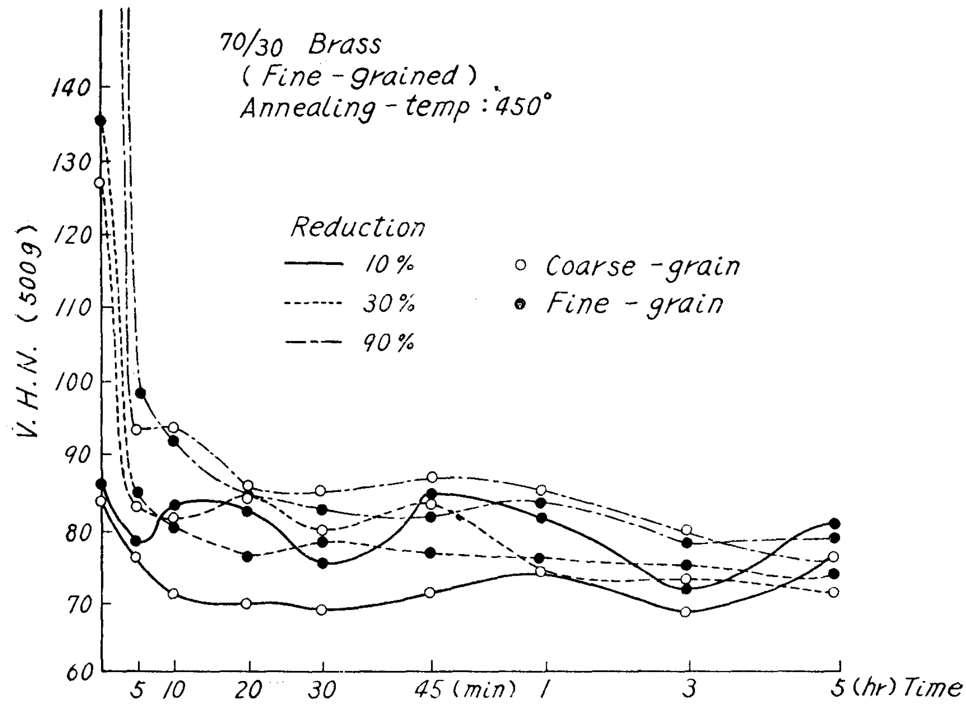


Fig. 8. Change in hardness at 450° of cold-rolled 70/30 brass.

tencency of softening, it was difficult to discriminate microscopically new grains, but slip-lines disappeared after annealing. Above-stated such discontinuous softening as in the case of 350° was hardly observable at 450°.

5. Annealing effect in pure copper

For comparison, the hardness of pure electrolytic copper was measured. The specimen was, after vacuum-melting, cold-rolled (0.5 mm thick) by 50 and 88 per cent. The hardness was measured after annealings at 150° and 250°. The results are shown in Fig. 9. At 250° the specimens of both reductions began to recrystallize at an early stage of annealing and no distinguishing feature was observable between them, while in the annealing at 150° the specimen of 50 per cent reduction showed discontinuity in the softening process. It is of interesting problem to study further on whether or not this phenomenon is due to the same mechanism as that in the above dicontinuous hardness change at 350° of cold-rolled brass.

Thus, the isothermal change in the hardness at the temperatures ranging from 150 to 450° was examined with regard to the various specimens different in ready-to-finish grain-size and working reduction, and it was clarified from the results that the ready-to-finish grain sizes or the development degree of the working structure had a considerable influence upon the change in hardness and its rate. As stated above, the hardness change does not show such a simple relationship between the anneal-hardening and the grain-size as hitherto reported[(1),(2),(3)],

(1) R.A. Wilkins and E. E. Bunn, *Copper Base Alloys*, (1943), 67 ; 202.
(2) Kato, Nishikawa and Suyama, J. Japan Inst. Met., **20** (1956), 234.
(3) Shinoda and Amano, ibid., **21** (1957), 62.

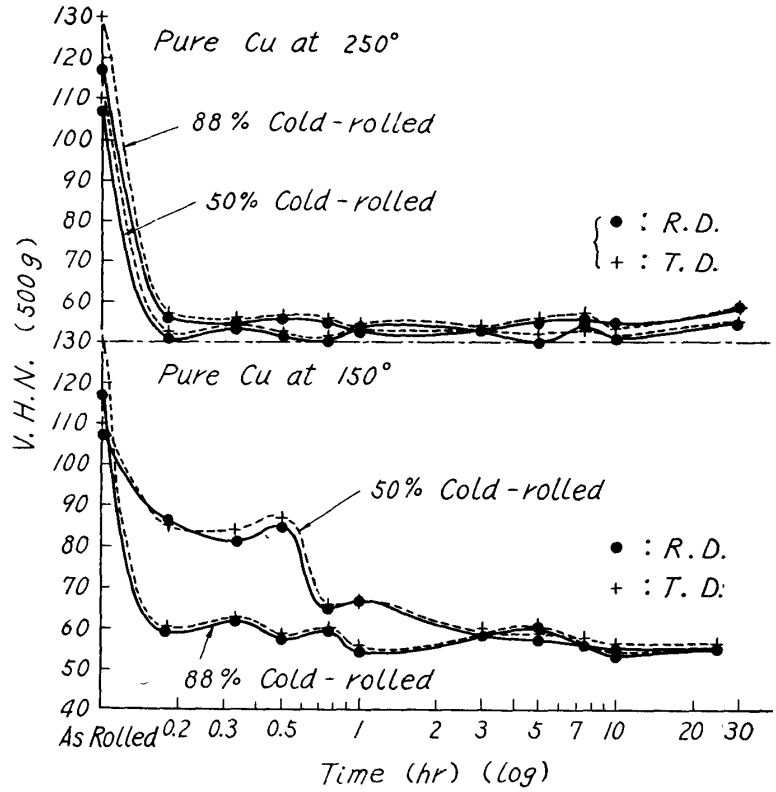


Fig. 9. Change in hardness at 150° and 250° of cold-rolled pure copper.

but is more complicatedly influenced by the development degree of the working structure. Such is also the case with the velocity of softening in the recrystallization process. The mechanism of anneal-hardening is not yet ascertained essencially, but it will be inferred from the hardness change at 150° that the primary hardening is influenced by the redistribution of the residual stress, while the secondary hardening by the behavior of the solute atoms in the material.

## Summary

The hardness change was measured when cold-rolled 70/30 brass sheets different in ready-to-finish grain size were annealed at the temperatures 150°~450°, after rolling of 10~90 per cent in reduction. The chief results may be summarized as follows:

(1) The hardness change is influenced by the ready-to-finish grain size, and accordingly, by the development degree of the working structure after cold-rolling.

(2) The primary hardening at 150° is not neccessarily in proportion to the working reduction, and tendencies are classified by the ready-to-finish grain sizes. From these facts, it is inferred that the primary hardening is affected by the degree of development and uniformity of the working strain.

(3) The secondary hardening at 150° increases with the increase of the working reduction, except the range of low reduction. The hardness change tends to be more influenced by the conditions of specimen such as compositions or impurities

rather than the grain size.

(4) In the highly reduced material, the anneal-hardening at 250° is smaller than that at 150°, but increases with the working reduction. In coarse-grined material, the recrystallization process is more retarded than in the fine-grained one.

(5) The recrystallization softening at 350° is not continuous, and after softening at the initial stage comes once hardening and thereafter again softening. In the reduction above 50 per cent, the amount of softening at an early stage still remains almost unchanged, but the hardening thereafter shows the maximum value on the way in proportion to the increase of working reduction. The working reduction at which the hardening above-mentioned shows maximum varies with the grain size.

(6) At 450° such discontinuous softening is hardly observable, but in coarse-grained material the velocity of recrystallization is also delayed.

(7) In the experiment on pure copper, at an early stage of annealing at 150° of the material of 50 per cent in reduction a slight delay in softening is observed, but it is not clear whether this can be connected with the change at 350° of cold-rolled brass.