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Recrystallization of 70/30 brass during induction heating

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The influence of the heating rate on the recrystallization kinetics and the as-recrystallized grain size has been studied for the case of heavily cold-drawn 70/30 brass. Samples have been continually reheated in muffle and induction furnaces to give heating rates between 0.18 and 180° C $\sec^{-1}$ and the progress of recrystallization followed by hardness and microscopy techniques. Recrystallization started at about 240° C and finished at about 320° C at the slowest heating rate: these temperatures were increased to about 340° C and 380° C, respectively, when the heating rate was increased to 180° C $\sec^{-1}$. At the same time, the hardness measurements indicated that the as-recrystallized grain size decreases from about 1.4 μm to about 0.8 μm. The results are discussed in relation to the normal recrystallization heat-treatments and the possibility of obtaining a temporary superplastic behaviour.

1. Introduction

There is an increasing trend to use rapid heating techniques in the heat-treatment field, either as part of a classic heat-treatment process (e.g. rapid austenitization to reduce grain growth) or in recrystallization annealing. In the latter case, rapid reheating should increase the temperature range for recrystallization leading to a more copious nucleation and hence a finer grain size. The techniques involved include induction heating, infra-red heating, modern jet gas ovens [1] and "continuous reheaters" where a drawn wire product forms the secondary coil of a transformer [2]. Such methods are preferable to the more classic salt baths since they are cleaner and safer and they are more easily applicable to continuous processes such as strip rolling or wire drawing. Furthermore, a continuous reheating process should give more uniform results than batch annealing (of a coil, for example).

However, in spite of the possible industrial interest in these techniques, little basic research appears to have been published on the influence of the heating rate on the metallurgical structure, with the exception of work by Naether *et al.* [3], Pavlidis *et al.* [4], Blanter *et al.* [5] and Zimin [6]. However, with the exception of Blanter *et al.*, the materials have contained precipitated impurities or second phases which interfered with the true recrystallization kinetics.

The present work has been limited to a study of the heating rate on the recrystallization kinetics with the principal aim of obtaining the finest possible grain size. The interest in the latter is two-fold: (a) an ultrafine grain size enhances the mechanical properties such as the yield, fracture and fatigue strengths [7]; (b) ultrafine grain-size materials usually behave superplastically, the permitted strain-rate range increasing strongly with decreasing grain size [8, 9]. Although a very fine grain structure is very unstable, imaginative process design should allow advantage to be taken of the temporary superplastic properties that it possesses to carry out high strain rate, low-energy consuming forming operations [10]. Furthermore, it is considered that the present comparison of conventional and rapid reheating should be of interest when applying recrystallization data obtained on small laboratory samples to the heat treatment of industrial components.

The material chosen for the present work was a standard 70/30 brass since it remains single phase

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TABLE I Analysis

Cu	Sn	Fe	Al	Ni	Zn
70.52 wt%	100 ppm	70 ppm	< 20 ppm	< 10 ppm	balance

at all temperatures and because its classical recrystallization behaviour has been widely studied [11–14]. In particular, Lamotte and Herenguel [14] have noted that, after 99.53% cold-rolling of a 70/30 brass, the structure consisted of imperfect subgrains of size 0.1 to 0.5 μm. On the other hand, Turley [15] found that, after 90 to 99% cold-rolling, no subgrain structure was observed, although electron diffraction patterns indicated the presence of a fine subgrain structure. He also found that heating for 30 min at 300° C led to small polygonized subgrains of about 0.05 μm diameter in regions of heavy deformation. Although it is clear that subsequent recrystallization treatments could not give a grain size smaller than this, it is not clear whether the grain size could even approach this lower limit. It is considered that induction heating would allow the nearest possible approach to this limit.

2. Experimental procedure

The 70/30 brass used in the present work was of a typical industrial quality and had the composition shown in Table I.

The conventionally cast ingot was extruded by Trefimetaux (billet temperature 750° C, container temperature 420° C, extrusion ratio 35) to 22 mm diameter bar having a grain size of 250 μm (Afnor NF A04 102 comparison method). The extruded product was then cold-drawn to 4 mm diameter wire (96.7% reduction). Samples were reheated in an induction furnace at various rates (55, 110, 180° C sec^{-1} with an accuracy of 5%) and water-quenched from different temperatures without hold periods. The quenching rate obtained was greater than 500° C sec^{-1}. The temperature was measured with a surface-welded thermocouple situated centrally in the heating zone. Calibration checks of such thermocouples showed them to lie easily within the normally accepted $\frac{3}{4}$% accuracy limits for chromel–alumel thermocouples. A series of specimens was also heated in a conventional muffle furnace to give a heating rate of 0.18° C sec^{-1}.

The heat-treated specimens were mounted and sectioned longitudinally. After mechanical polishing, the specimens were electropolished in Struers D31 solution at 12 V and etched in alcoholic ferric chloride (50 g $FeCl_3$, 150 ml HCl, 600 ml C_2H_5OH). The progress of recrystallization was followed by optical microscopy and microhardness tests which were carried out using a Leitz microhardness tester with a 200 g load. The average of at least ten impressions was taken for each specimen. When it was not too small, the grain size was measured from enlarged optical micrographs using the mean linear intercept method. Limited electron microscopy was carried out using conventional thin foil techniques.

3. Results and discussion

3.1. The cold-worked state

The as-received cold-worked wire showed bands on longitudinal sections (Fig. 1a) and "islands" on transverse sections (Fig. 1b) which etched less

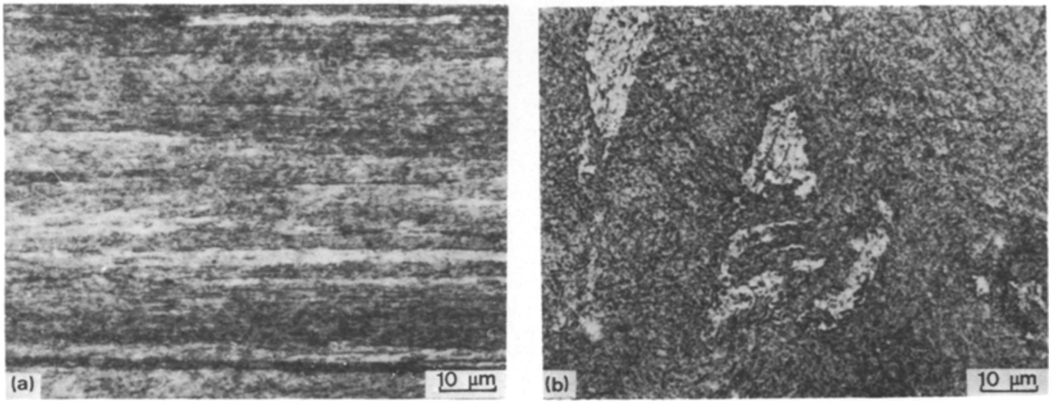

Figure 1 Microstructural heterogeneity in the as-received 96.7% cold-drawn wire. (a) Longitudinal section. (b) Transverse section.

readily. These regions were too small to allow meaningful microhardness measurements to be made. Work by Whitwham *et al.* [13], Lamotte and Herenguel [14], Turley [15] Cook and Richards [16], Herenguel *et al.* [17] and Blanchard *et al.* [18] has clearly shown that, following heavy cold-rolling of brass, the structure consists of trapezoidal or lens-shaped regions of material work-hardened by slip in a heavily sheared matrix which etches more easily, and that the boundaries between the lightly and heavily deformed regions make a constant angle of ± 35° with the sheet surface. It is considered that the differently etching regions found in the present work, although having a different form due to the deformation process used, are nevertheless due to a heterogeneous deformation on a microscopic scale.

3.2. Microhardness measurements

As noted above, the progress of structural changes during reheating was followed by microhardness measurements. Preliminary tests showed that even surface welding the thermocouples to the samples could have a major influence on the structure, leading to recrystallization and grain growth in the heat affected zone (HAZ; Fig. 2a). Fig. 2b compares thickness hardness traverses adjacent to the weld and far from the weld and indicates an HAZ to a depth of 1 mm. Although the hardness measurements agree close to the centre, the possible errors introduced by the HAZ were avoided by longitudinally displacing the hardness measurements several millimeters from the weld zone. The justification for this is shown in Fig. 3 which shows the longitudinal hardness gradient at the centre of a specimen which was induction heated without a thermocouple. It can be seen that the length of uniform hardness (and therefore of uniform structure), although not equal to the induction coil length, is nevertheless large enough to allow the hardness measurements to be made a reasonable distance from the centrally located thermocouple. This problem can only be overcome by use of a pyrometer measuring system but this introduces further errors due to the lack of knowledge regarding the change of emissivity with time and temperature. It is also of interest to note that the chosen induction coil spacing did not lead to any periodic structural variations.

3.3. Progress of recrystallization

The change in mean Vickers hardness is shown as a function of heating time (i.e. time from the start of heating to start of quench) in Fig. 4 for the conventionally and induction-heated samples. In all cases, the hardness increases from the cold-worked state to a maximum, followed by the characteristic rapid decrease due to recrystallization, followed by a slower decrease due to grain growth.

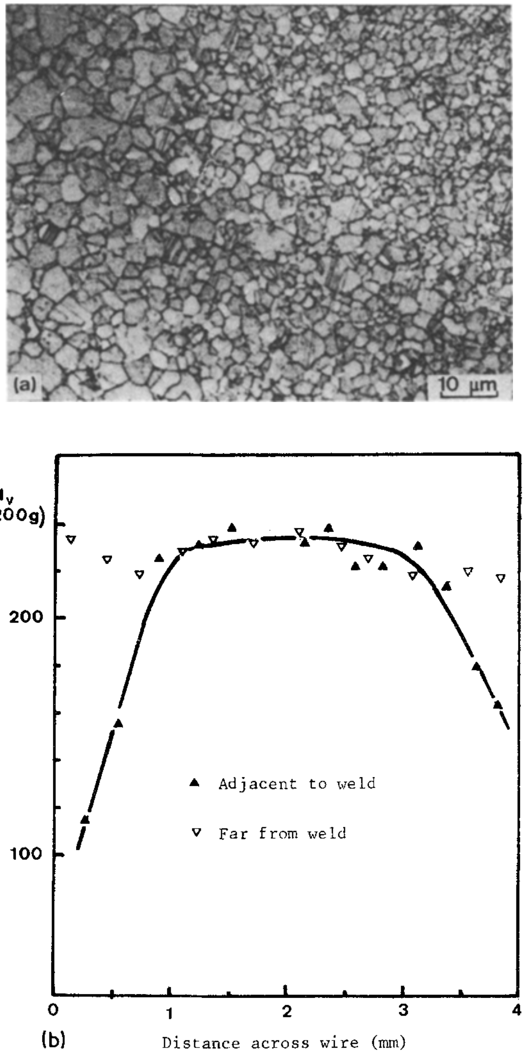

Figure 2 (a) Typical structural gradient close to weld. (b) Typical through-thickness microhardness traverses in heat-treated wire adjacent to and far from thermocouple weld position (3 sec at 110° C sec^{-1}).

The initial hardness increase, which was not accompanied by any optically observable structural changes, is well established and has been attributed to a redistribution of internal stresses [19] or a restoration of short-range order [20, 21]. The maximum hardness appears to be independent of

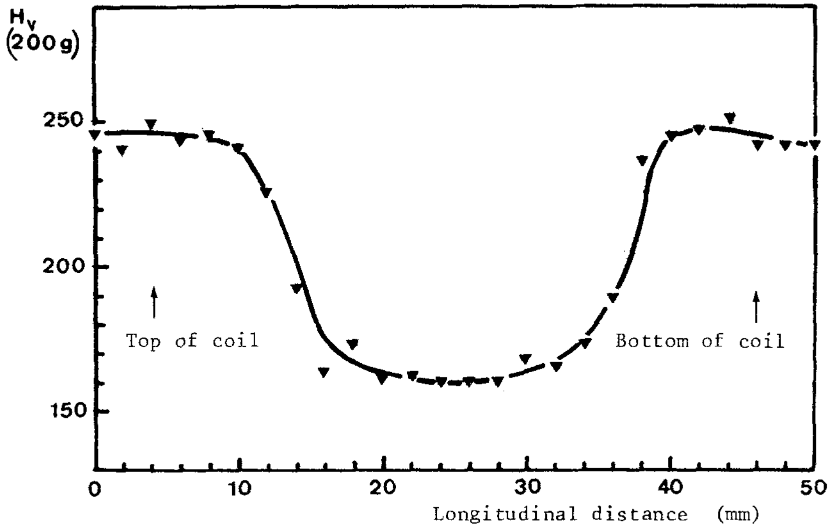

Figure 3 Longitudinal variation of Vickers hardness at centre of induction heated wire (2.3 sec at 180° C sec^{-1}). The arrows indicate the outside edges of the coil.

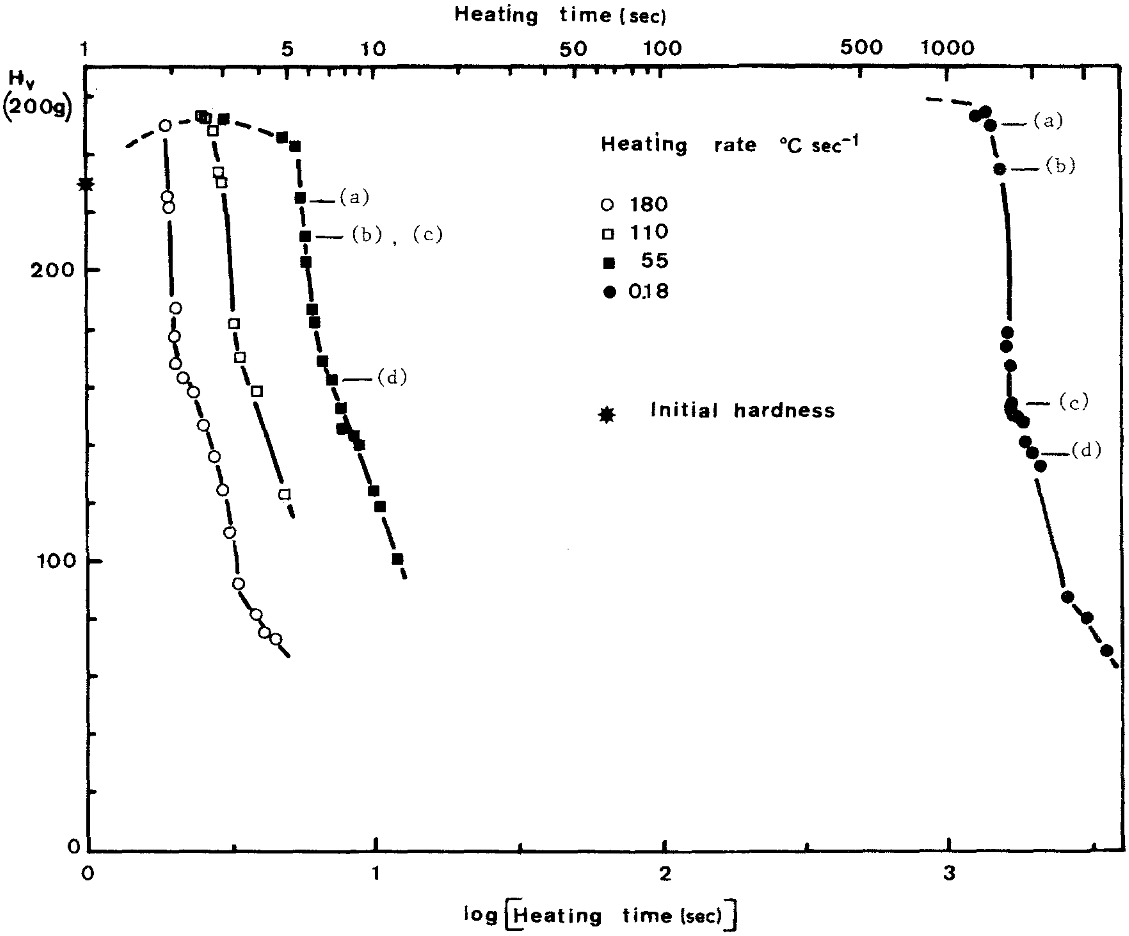

Figure 4 Change in mean Vickers hardness as a function of time for induction and conventionally heated samples. The letters adjacent to points on the 0.18 and 55° C sec^{-1} curves refer to micrographs in Figs. 6 and 7, respectively.

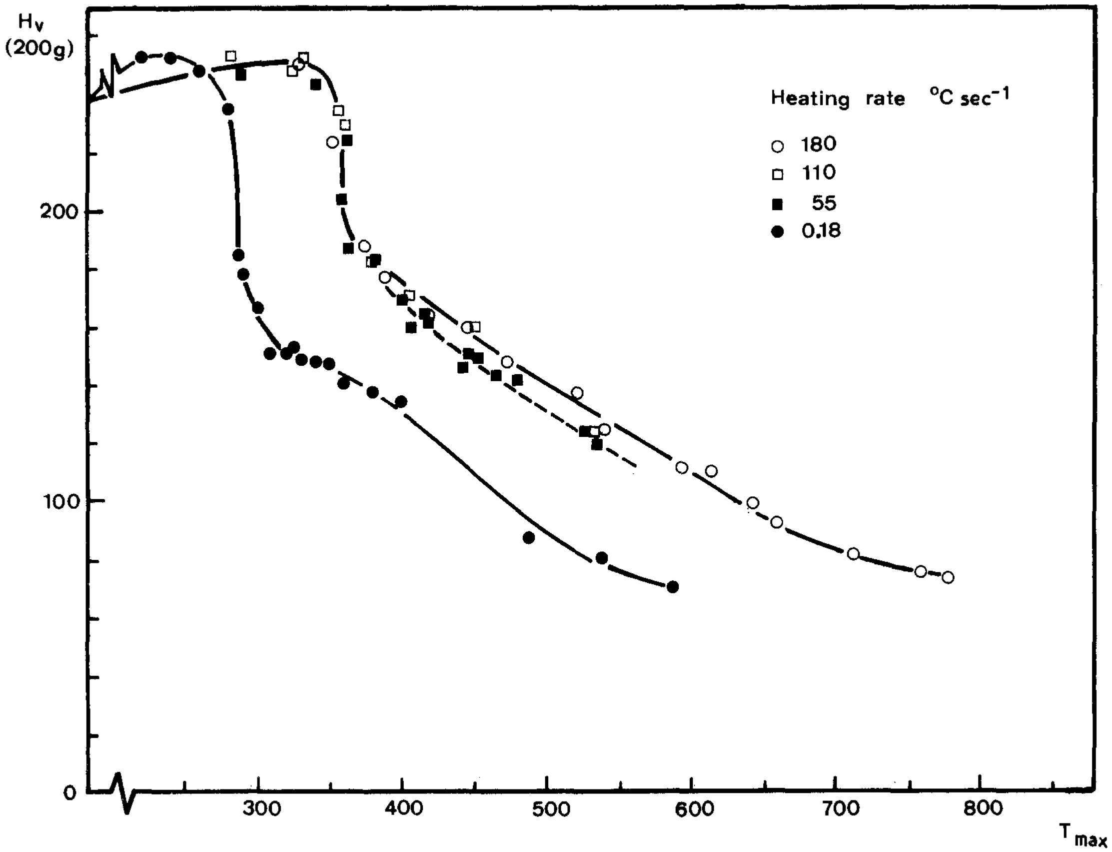

Figure 5 Change in mean Vickers hardness as a function of the maximum temperature for conventionally and induction-heated samples.

the heating rate. Fig. 4 shows that, as expected, recrystallization starts and finishes at earlier times as the heating rate increases. However, when the hardness data of induction-heated samples are plotted as a function of the maximum temperature $T_{\max}$ (i.e. the temperature prior to quenching; Fig. 5), it can be seen that, within the present experimental accuracy, both recrystallization and grain growth appear independent of the heating rate in the 55 to 180° C sec^{-1} range, although a slight effect of heating rate may be distinguished in the grain-growth region, as indicated by the broken line. Fig. 5 also shows that when a heating rate of 0.18° C sec^{-1} is used (i.e. 10^3 times slower), recrystallization starts and finishes at 60 to 80° C lower. Furthermore, the temperature range for recrystallization appears to be greater for the slowest heating rate. In addition, it appears that the grain-growth rate is slightly faster for the very slow heating rate, thereby agreeing with previous work which has established that both time and temperature influence grain growth.

Thus, it appears that the recrystallization temperature increases with an increasing heating rate. This is not unexpected since recrystallization is both a temperature and time-dependent process. However, work by Zimin [6] showed a decrease in the recrystallization temperature of a 38 wt% Zn brass from 430° C at 100° C sec^{-1} to 395° C at 400° C sec^{-1} but further increases in the heating rate above 450° C sec^{-1} led to increases in the recrystallization temperature. However, this alloy was two-phase which clearly complicates the situation, particularly in view of the fact that the b c c β' phase partially undergoes a martensitic transformation to a supersaturated f c c α phase during straining prior to recrystallization. It is more valid to compare the present work with that of Blanter *et al.* [5] who found a gradual increase in the recrystallization temperature of a single-phase 32 wt% Zn brass from 350 to 450° C when the heating rate was increased from 0.11 to 500° C sec^{-1}. Although their data are not clearly presented and their prior deformation was only 50%, they clearly found the same trend and the same order of magnitude in the change of recrys-

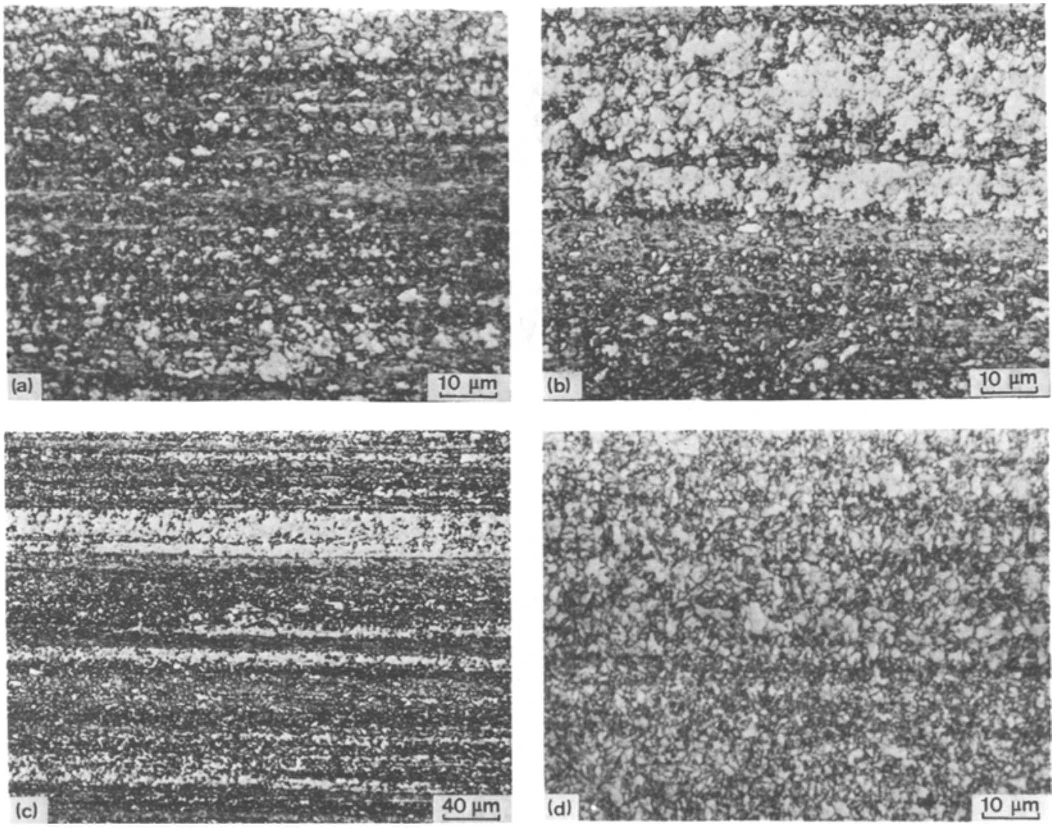

Figure 6 Structural changes during the slow reheating (0.18° C $\sec^{-1}$) of heavily cold-worked brass. (a) Heating time 22 min, $T_{\max}$ 260° C. (b) Heating time 24 min, $T_{\max}$ 280° C. (c) Heating time 28.2 min, $T_{\max}$ 326° C. (d) Heating time 33.1 min, $T_{\max}$ 380° C.

tallization temperature with heating rate as in the present work. Thus, from the hardness versus final temperature curves, it may be concluded that, within the range of normal induction heating, the heating rate has little or no effect on the final hardness at a given final temperature.

We will now consider the hardness curves (Figs. 4 and 5), in greater detail, particularly in regard to the end of recrystallization. As noted above, previous work [17, 19] has shown that, after heavy cold-rolling, 70/30 brass is heterogeneously deformed. The same workers have shown that, during subsequent reheating, recrystallization occurs firstly in the regions of heavy shear, leading to a grain size of 2 to 3 μm, this change coinciding with the rapid hardness drop. The softening rate then decreases and is associated with a delayed recrystallization in the less heavily deformed zones to give a grain size two or three times greater than that of the first family. This stage is then followed by one of relative stability during which little growth occurs and finally growth occurs more rapidly with the second family of grains growing at the expense of the first family. Thus, it appears that, at least for the case of heavily rolled 70/30 brass, the structural changes are more complicated than the normally accepted single-step recrystallization followed by grain growth.

Considering first the slowly heated samples, Fig. 4 and especially Fig. 5 show the same type of hardness changes as discussed above, with the initial rapid drop due to recrystallization being followed by an arrest in the curve, followed by a decrease due to grain growth. The final levelling off of the hardness curve may be due to either a reduction in the growth rate or due to the fact that, at larger grain sizes, the hardness impression approaches the grain size, thereby restricting the validity of the former.

Optical microscopy clearly shows the appearance of the recrystallized grains in the cold-work matrix (Fig. 6a, b) but did not allow the end of recrystallization to be defined clearly. For example,

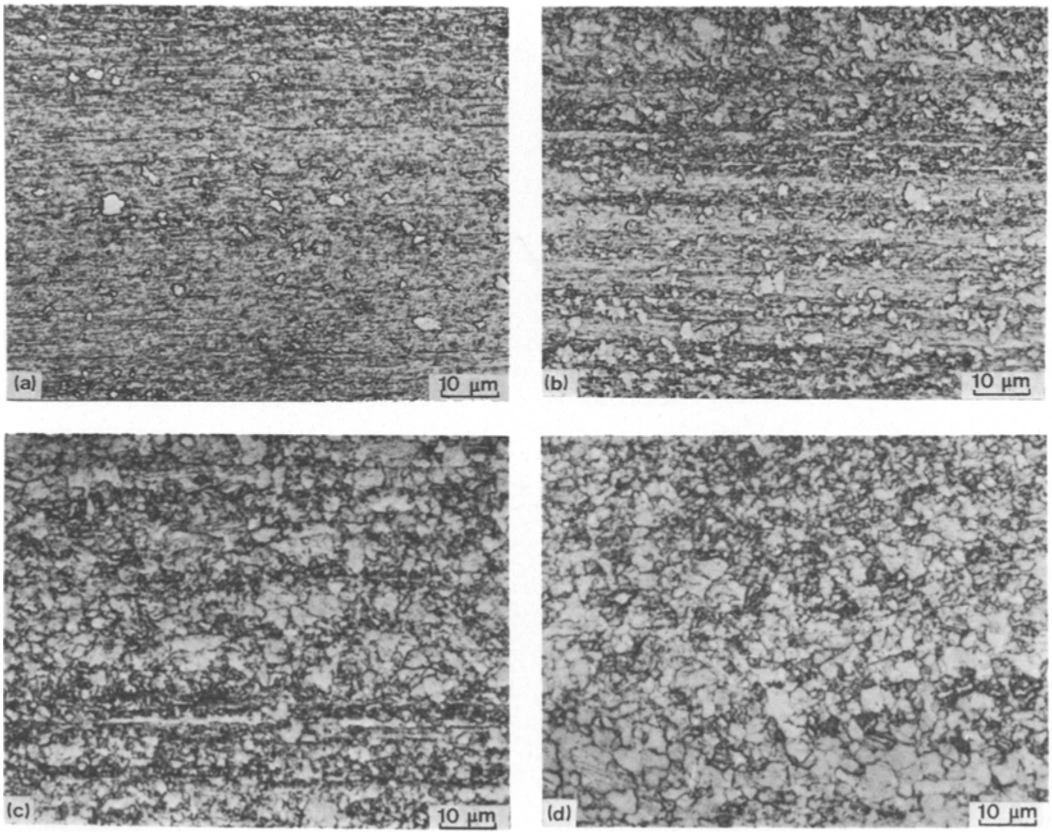

Figure 7 Structural changes during the induction heating (55° C $\sec^{-1}$) of heavily cold-worked brass. (a) Heating time 5.6 sec, $T_{\max}$ 330° C. (b) Heating time 5.9 sec, $T_{\max}$ 345° C. (c) Heating time 5.9 sec, $T_{\max}$ 345° C. (d) Heating time 7.1 sec, $T_{\max}$ 410° C.

a highly recrystallized structure but with areas of darker contrast (Fig. 6c) was found near the end of the rapid hardness drop. It was unclear whether the darker areas were due to remnants of the cold-worked structure or regions of extremely fine grains. This same uncertainty is still present (Fig. 6d), even when the hardness curves would tend to indicate 100% recrystallization. In spite of the uncertainty regarding the end of recrystallization, it would appear that the latter occurs at a Vickers hardness of 150 ± 2.

Considering the induction-heated samples, Fig. 4 does not indicate any clear arrest at the completion of recrystallization. The hardness changes as a function of $T_{\max}$ (Fig. 5), again indicate an apparent lack of arrest after the major hardness decrease, this being particularly striking when compared with the curve for the heating rate of 0.18° C $\sec^{-1}$. It therefore appears that the two-stage recrystallization process does not occur during induction heating or, if it does, the two stages are not resolvable by the techniques used in the present work. Optical micrographs at various stages of softening for the samples heated at 55° C $\sec^{-1}$ are given in Fig. 7. As before, the development of recrystallization is clearly shown (Fig. 7a, b) although the process occurs heterogeneously at early stages (Fig. 7c). In later stages, it was again difficult to distinguish between very small submicron recrystallized grains and the remnants of a cold-worked structure (Fig. 7d). However, the combination of metallography and hardness testing indicated a hardness of 180 ± 5 at the end of recrystallization for all the induction heating rates.

3.4. Grain-size determination

In view of the difficulties of measuring the extremely fine grain sizes obtained, it was decided to deduce the recrystallized grain sizes from the hardness measurements. Whitwham *et al.* [13] have verified the empirical relation of Basset and Davis [22] between the Brinell hardness, H_B, and the mean grain diameter, $\bar{L}$ (μm): $H_B = 168.6$

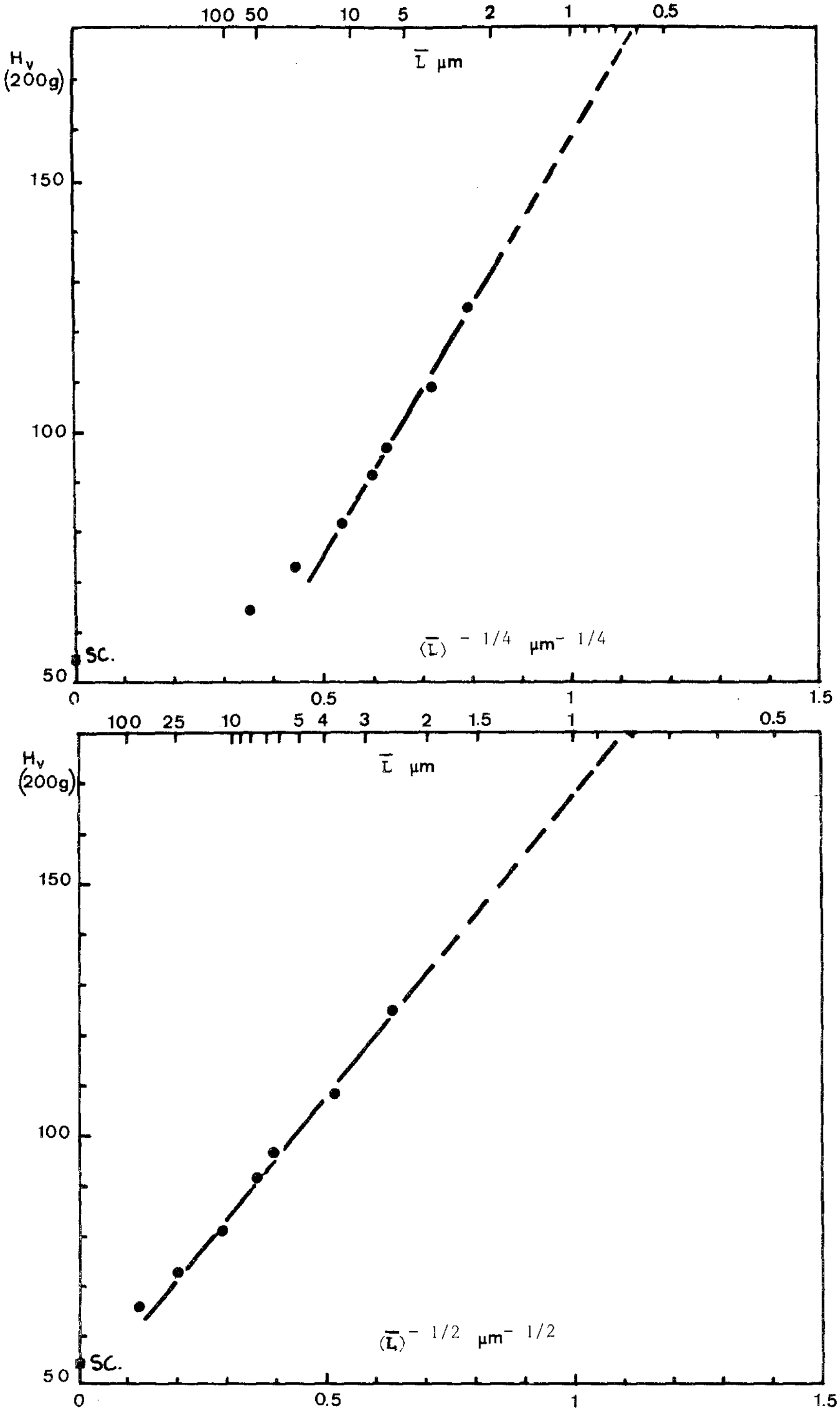

Figure 8 Relations between the mean Vickers hardness (200 g) and the grain size (SC = single crystal).

$(\bar{L})^{-1/4}$. They note that this relation was valid for 70/30 brass (amongst other alloys) down to their minimum grain size of 2 μm. Later work [19] demonstrated the validity of this $(\bar{L})^{-1/4}$ law for grains or subgrains in aluminium down to their minimum values of about 1 μm. On the other hand, Jindal and Armstrong [23] and Armstrong [24] have shown from their work and that of others that the grain-size dependence of the hardness follows a Hall–Petch type equation of the form: $H = H_0 + K(\bar{L})^{-1/2}$. Hence there appears to be an ambiguity whether the hardness depends on the inverse of the square or fourth root of the grain size, although the former would appear more fundamental. However, it should be noted that the two relations are very similar for low $\bar{L}$ values. When values of the mean microhardness obtained in the present work were plotted as a function of either $(\bar{L})^{-1/2}$ or $(\bar{L})^{-1/4}$ ($\bar{L}$ being the mean linear intercept grain size in μm), straight lines were obtained in both cases, except for grain sizes greater than about 25 μm, when the diamond impression and grain size approach each other. The two equations obtained were:

$$H_V = 47 + 118\,(\bar{L})^{-1/2}$$

$$H_V = -10 + 168\,(\bar{L})^{-1/4}.$$

These curves are plotted in Fig. 8 together with the measured values. Hence the present work does not clarify the above noted ambiguity but it is clear that both forms of the equation are valid over the limited grain-size range encountered in the present work. Assuming that it is valid to extrapolate the $(\bar{L})^{-1/2}$ curve to smaller grain sizes, the above noted hardness measurements at the end of recrystallization then give mean intercept grain sizes of 0.80 ± 0.05 and 1.40 ± 0.05 μm for the induction and conventionally heated samples, respectively (the $(\bar{L})^{-1/4}$ equation gives slightly smaller values: 0.60 ± 0.05 and 1.20 ± 0.05 μm, respectively). The limited electron microscopy carried out confirmed that these values are essentially correct, and tended to agree with the lower values deduced from the hardness measurements.

Thus a 1000-fold increase in the heating rate (from 0.18 to 180° C sec^{-1}) leads to only a decrease from 1.4μm to 0.8μm in the recrystallized grain size and an increase from about 320° C to about 380° C in the temperature for 100% recrystallization. Hence, it is not surprising that the present range of induction heating rates (a ratio of 3.3) gave no detectable shifts in either grain size or recrystallization temperature. To produce a further markedly increased recrystallization temperature and decreased grain size, it would be necessary to increase the heating rate by such an amount that it would not be attainable (especially industrially). It should also be noted that the as-recrystallized hardness cannot be a linear function of the heating rate since, otherwise, an infinite heating rate would produce a hardness superior to the as-cold-worked hardness.

Hence, for all practical purposes, the above noted grain size of 0.8 or 0.6 μm would appear to be the minimum attainable for the present material conditions. There would appear to be two variables which could further reduce this value. Firstly, the recrystallized grain size decreases with increasing prior deformation. However, the reduction used in the present work (96.7%) is already extremely high, and previous work [12] indicates that a further increase would have a negligible effect. Secondly, a finer initial grain size would be expected to lead to a finer recrystallized grain size. Although this is true for low prior deformations, the work of Pulsifier [12] clearly indicates that the influence of the initial grain size is almost negligible at the high reduction used in the present work. However, the influence of these two factors is such that it appears likely that a lower prior deformation (which is more feasible industrially) could be compensated for by a smaller initial grain size.

4. Applications

At present, many recrystallization treatments are carried out by heating to, and holding at, a temperature low in the recrystallization range. This is not conducive to a high nucleation rate and, therefore, the grain size obtained is not the finest possible. Metallurgically speaking, it is more favourable to heat to a higher temperature (corresponding to the end of recrystallization) without an isothermal hold; this temperature will depend on the heating rate used which should be the highest possible compatible with a uniform temperature distribution in the product. In addition, depending on the production conditions, it may well be economically advantageous to offset the slightly higher furnace temperature by the greatly increased throughput resulting from the absence of a hold time.

Finally, it is of interest to consider the possible consequences of rapid reheating on the temporary superplastic behaviour. Superplasticity usually occurs at temperatures greater than half the absolute melting temperature, i.e. greater than 340° C for a 70/30 brass. Fig. 5 clearly shows that, in the case of slow reheating (0.18° C sec^{-1}), grain growth is already beginning to occur at this temperature, whereas, in the case of induction heating, recrystallization has hardly started. Hence rapid reheating should be beneficial to obtaining a superplastic behaviour since the as-recrystallized grain size is finer and recrystallization is delayed to a higher temperature. This work is currently being carried out.

5. Conclusions

During the continual reheating of a heavily cold-worked 70/30 brass, the temperature for the start of recrystallization increases from 240 to 340° C and that for the end of recrystallization from 320 to 380° C when the heating rate is increased from 0.18 to 180° $\mathrm{C\,sec^{-1}}$. This increase in recrystallization temperature is accompanied by a decrease in the as-recrystallized grain size from about 1.4 μm to about 0.8 μm (evaluated by microhardness measurements).

A relatively small change in the induction heating rate (from 55° $\mathrm{C\,sec^{-1}}$ to 180° $\mathrm{C\,sec^{-1}}$) has no measurable influence on the recrystallization kinetics or the as-recrystallized grain size.

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