

EFFECT OF ZINC CONTENT ON THE ANNEALING BEHAVIOUR OF DEFORMED ALPHA-BRASSES

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Hardness measurements, metallographic observations, as well as X-ray diffraction patterns were used to study the annealing of deformed four alpha-brasses of zinc content 3.5, 8.8, 19.4 and 26.8 weight percent. The relaxation spectrum for each alloy was characterized by the presence of three annealing stages. The first one was attributed to the annealing out of vacancies and relative free dislocations. The second annealing stage, observed at higher annealing temperature, was attributed to recrystallization mechanism which takes place by self-diffusion. At much higher temperatures, grain growth mechanism was responsible for the third annealing stage. Increasing zinc content was found to help the annealing mechanisms, as it increases the rate of vacancy diffusion in the matrix, moreover it facilitates slip reactions. This increase takes place in two rates, high rate in alloys of low zinc content, and relative lower rate in alloys of high zinc content.

1. Introduction

In previous work^{1,2)} one of the authors (T. H. Youssef), pointed towards the great effect of zinc content on the plastic properties of alpha-brasses.

Since vacancies and dislocations play an important role in deformed matrices, it was thought worth-while to investigate the annealing kinetics for the

deformed alpha-brasses and to study the influence of the interaction of solute-zinc atoms with present defects.

The alpha-brasses were chosen for this study because the dislocation structure produced by deformation range from intergranular cells in the low zinc content alloys to planar pile ups against obstacles in the high zinc brasses³⁾.

2. Experimental work and results

Samples of Cu-Zn alloys studied: 3.5, 8.8, 19.4 and 26.8 weight % Zn, were prepared by the method previously described⁴⁾. The prepared specimens were in the form of sheets of $20 \times 10 \times 0.2$ mm. All the specimens were annealed at 200°C for 10 hours to bring them to a common standard conditions. Then they were cold-drawn to 25% tension. Annealing of the samples was carried-out in a silica tube evacuated to 10^{-3} Pa, using a furnace provided with a thermoregulator in which the temperature could be maintained constant for several hours to within $\pm 2^\circ\text{C}$. The vickers hardness number, V. H. N., of the annealed samples was measured at room temperature (25°C) with a hardness tester WPM (Leipzig). An average of at least five different observation runs was taken.

Isothermal annealing runs relating vickers hardness measurements at increasing temperature ranges were done. Typical sets of observations showing the change in relative hardness value, $(\Delta H/H_0)$, versus the annealing time, t , at different temperatures (range between 100°C and 625°C), are represented in Fig. 1, for the four studied alloys, respectively. Here (ΔH) is equal to $(H_0 - H')$; as (H_0) is the hardness of the deformed sample before annealing, (H') is the hardness after isothermal annealing for time t .

From Fig. 1 it is possible to note:

- (a) The annealing process goes on through 3 stages in each alloy.
- (b) The amount of recovery, as measured by the value $(\Delta H/H_0)$, for the same annealing temperature decreased with increasing zinc content. From the isothermal data of Fig. 1, the activation energy for each stage was calculated by making cuts at different values of $(\Delta H/H_0)$, for the four studied alloys. For each cut the logarithm of the annealing time required to reach that value was plotted against the corresponding reciprocal of absolute annealing temperature. Nearly parallel lines were obtained for each stage as shown in Fig. 2. The values of the calculated activation energies corresponding to the three annealing stages in the four studied alloys are reported in Table 1.

TABLE 1

Annealing stage	Activation energy E calculated in eV for			
	Cu-3.5% Zn	Cu-8.8% Zn	Cu-19.4% Zn	Cu-26.8% Zn
First stage	0.65	0.57	0.50	0.45
Second stage	1.40	1.20	1.00	0.90
Third stage	0.85	0.72	0.67	0.57

It is clear that increasing zinc content is associated with a decrease in the activation energy for each stage.

The associated microstructure variations due to annealing were followed after each annealing process by X-ray photographs using Cu K radiation, and by optical microscopic photographs. The results are shown in Fig. 3 taken at four states for each alloy studied, in which:

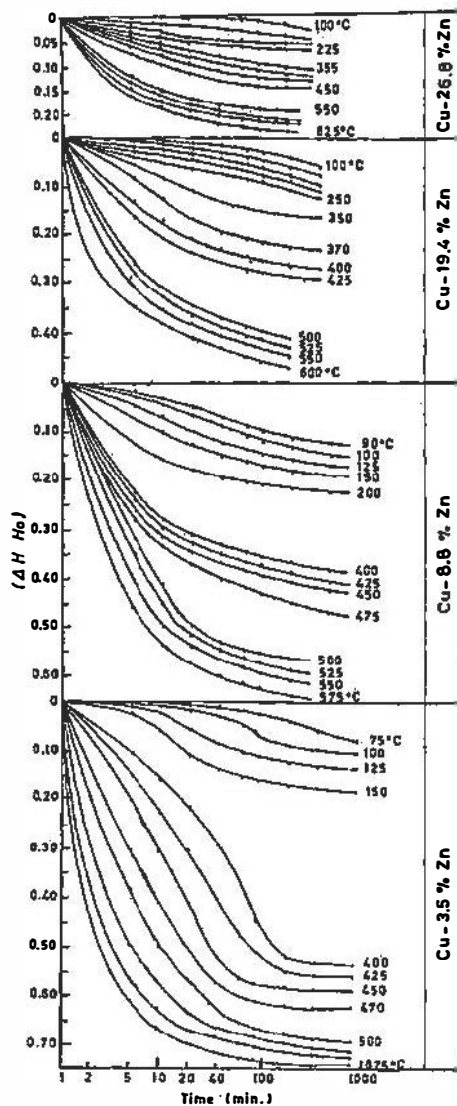


Fig. 1. Typical sets of isothermal annealing curves for the four alloys studied (3.5, 8.8, 19.4, 26.8 weight % Zn), showing the variation of $(\Delta H/H_0)$ with time at different annealing temperatures.

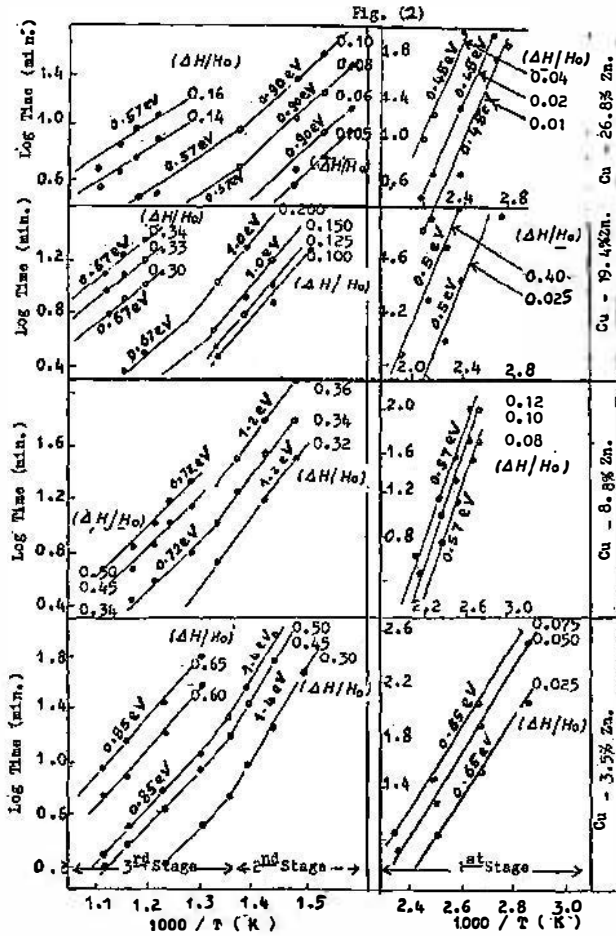


Fig. 2. Typical sets of curves showing times and temperatures for the four alloys (3.5, 8.8, 19.4, 26.8 weight % Zn) to calculate the energies activating the 3 annealing stages. Curves were derived from cuts taken from Fig. 1.

Section (A) represents deformed samples before annealing.

Section (B) represents samples annealed at 200 °C in which the X-ray pattern indicates that it is a recovery annealing stage.

Section (C) for samples annealed at 400°C, in which X-ray patterns exhibit continuous lines of maximum sharpness and the microstructure shows nucleation of small grains in the matrix indicating a recrystallization stage.

Section (D) for samples annealed at 600 °C in which growth of new grains and spotty Debye-Scherrer pattern indicates grain growth in this stage.

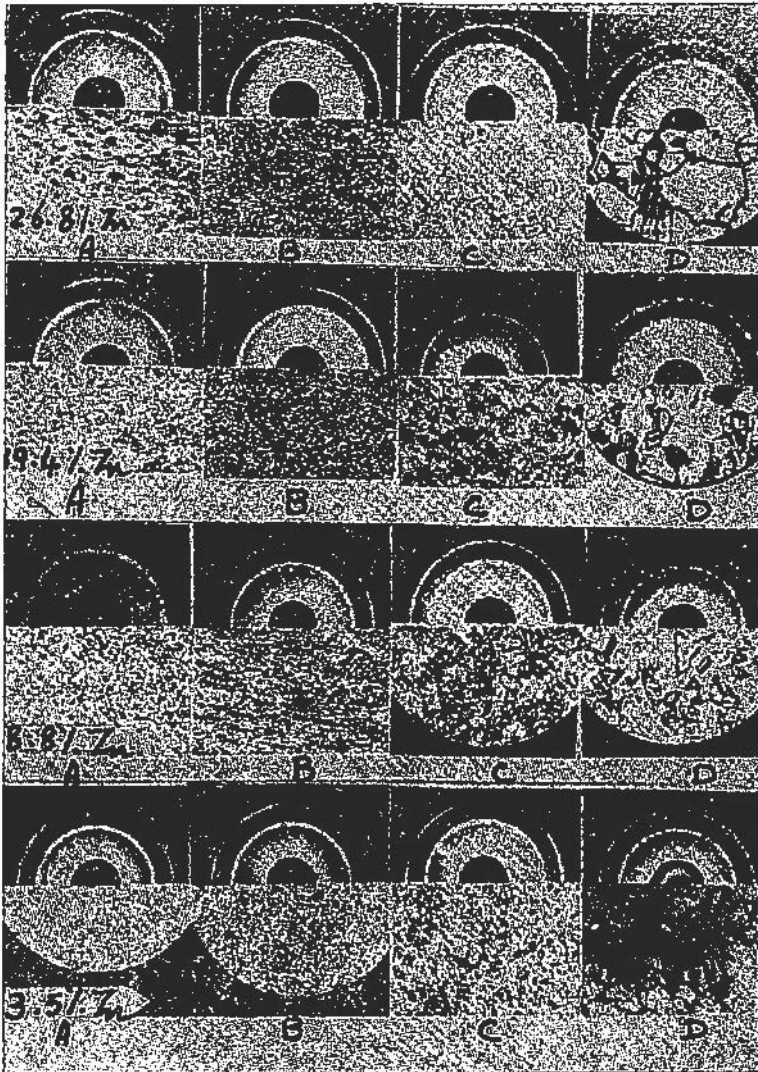


Fig. 3. X-ray diffraction and optical microstructures observed in the four alloys (3.5, 8.8, 19.4, 26.8 weight % Zn). Magnification X 200.

3. Discussion

It is generally accepted that a deformed matrix contains dislocations⁵⁾ distributed throughout producing internal strains⁶⁾. In addition to these dislocations, vacancies⁷⁾ are also found in amount considerably in excess of their equilibrium concentration. These vacancies will migrate in the matrix during recovery process with an activation energy will be trapped with zinc atoms forming vacancy-zinc

pairs due to the high vacancy affinity to zinc atoms. The association of vacancies and zinc atoms may be important in bringing an increase in the effective vacancy concentration near zinc atoms, so that the rate of diffusion will be increased⁹⁾. As thermal energy is supplied by annealing, these vacancy-zinc pairs are rendered mobile and migrate randomly in the matrix. The calculated activation energy for recovery in Cu-3.5% Zn alloy equals to 0.65 eV (as obtained from Fig. 2) seems to be that for migration of vacancy-zinc in this alloy, which is in agreement with similar results done before¹⁰⁾.

By increasing zinc content the activation energies corresponding to this stage for Cu-1.8% Zn, Cu-19.4% Zn, and Cu-26.8% Zn are 0.57, 0.50 and 0.45 eV, respectively. This decrease in activation energies might be due to the fact that increasing zinc content will be accompanied by increased number of vacancy-zinc pairs, and decrease in the number of absolute vacancies in the matrix; as a result the activation energy will be the resultant energy for migration of existing pure vacancies as well as vacancy-zinc pairs. The migration energy for pure vacancies is 0.76 eV^{8, 11)}, and that of vacancy-zinc pairs is 0.41¹²⁾. As a result increasing vacancy-zinc pairs will lead to a decrease in the net activation energy for recovery. The dependence of this decrease, (which is assumed to be due to existing vacancy-zinc pairs), on zinc content is given in Table 2.

TABLE 2.

Alloy	Activation energy E , for recovery in eV	Difference in E due to addition of zinc (D) $D = E_0 - E$	$D^2 = (E_0 - E)^2$
Pure copper	0.76 E_0	0	0
Cu-3.5% Zn	0.65	0.11	1.21×10^{-2}
Cu-8.8% Zn	0.57	0.19	3.61×10^{-2}
Cu-19.4% Zn	0.50	0.26	6.76×10^{-2}
Cu-26.8% Zn	0.45	0.31	9.61×10^{-2}

Fig. 4 shows that $(E_0 - E)^2$ increase linearly with zinc content which agrees with diffusion reactions.

In the second annealing stage the X -ray diffraction patterns, Fig. 3-C shows a continuous line of maximum sharpness, which shows that annealing in this stage depends on rearrangement of dislocations in the matrix¹³⁾. In alloys of high zinc content, small spotty rings were observed indicating the finishing of recrystallization reaction and earlier beginning of grain growth in this range due to increased zinc content⁹⁾. This difference in X -ray pattern might be due to difference in the deformed matrix; in alpha-brasses of low zinc content it contains cellular dislocation pattern, while it contains dislocation pile-ups against obstacles in alpha-brasses of high zinc content. The microstructures accompanying this stage show nucleation of small grains in the matrix.

The calculated activation energies corresponding to this stage for these studied alloys are 1.4, 1.2, 1.0, and 0.9 eV for the four studied alloys (3.5, 8.8, 19.4 and 26.8 weight % Zn), respectively. These values are found to energies for self diffu-

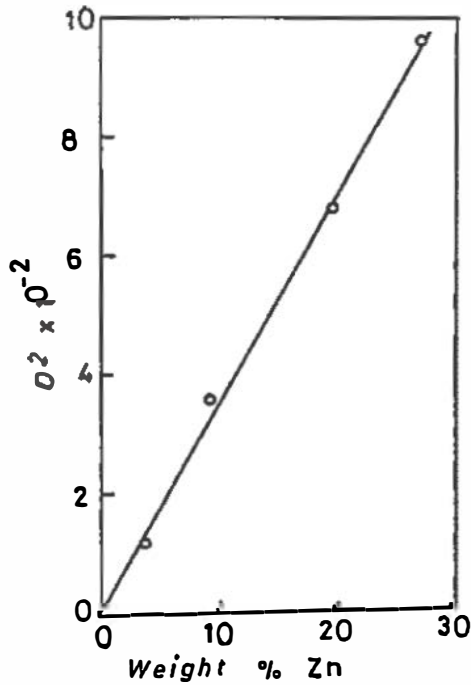


Fig. 4. Dependence of (decrease in activation energy)² on zinc content.

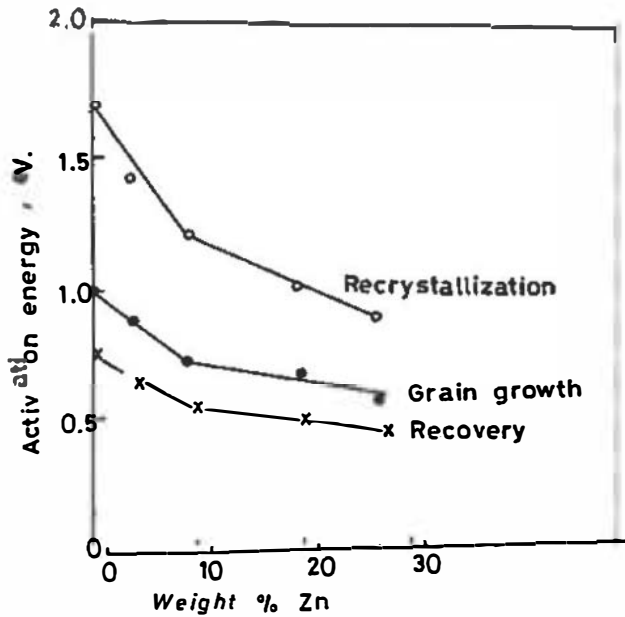


Fig. 5. Variation of activation energy (E) with zinc content for the three annealing stages.

sion in alpha-brasses¹³⁾. These values show that recrystallization proceeds more easily than those operative in pure copper (1.7 eV)¹⁵⁾. This might be because recrystallization mechanism depends on self-diffusion reaction which is enhanced by increasing zinc content⁹⁾.

As for the third annealing stage the X-ray diffraction pattern accompanied this stage show clearly the growth of new grains, and the spotty Laue pattern in the rings show itself. Also the microstructures accompanying this annealing stage show clearly the growth of the new grains. This is clearly shown in section D in Fig. 3. The calculated activation energies in this temperature range are found to be 0.85, 0.72, 0.67, and 0.57 eV for the four copper alloys containing 3.5, 8.8, 19.4, and 26.8 weight % Zn, respectively. These values of activation energies are in agreement with those previously calculated for grain growth in alpha-brasses¹⁶⁾ and agree with the suggestion put down by Mott¹⁷⁾ that grain boundary diffusion is the mechanism controlling grain growth in alpha-brasses. The decrease of activation energy for recovery, which shows that the rate of decrease of activation energy in the 3 mechanisms is higher in alloys with low zinc content than that in alloys with high zinc content. This behaviour indicates that annealing in low zinc alloys is easier than that in the highest zinc content. This might be because of the difference in the deformed matrix. In low zinc alloys the matrix contains intergranular cells while those of higher zinc contain planar pile ups against obstacles.

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UTJECAJ SADRŽAJA CINKA NA POPUŠTANJE DEFORMIRANE ALFA-MJEDI

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Popuštanje deformiranih četiriju uzoraka alfa-mjedi, s 3,5, 8,8, 19,4 i 26,8 tež. postotaka cinka, proučavano je mjerenjem tvrdoće, metalografskim zapažanjima i rendgenskim difrakcijskim slikama. Spektar oporavka svake slitine karakteriziran je s tri stupnja popuštanja. Prvi se pripisuje otpuštanju vakancija i relativno slobodnih dislokacija. Drugi stupanj popuštanja, zapažen pri višim temperaturama, pripisan je mehanizmu rekristalizacije koji se odvija samodifuzijom. Za treći stupanj popuštanja-žarenja pri znatno višim temperaturama, odgovoran je mehanizam rasta zrna. Utvrđeno je da veći sadržaj cinka pospješuje proces popuštanja, jer pojačava brzinu difuzije vakancija u maticu, a osim toga olakšava reakcije klizanja. Popuštanja u slitinama s niskim sadržajem cinka odvija se lakše od onog u slitinama s najvećim sadržajem cinka.