

Lattice Parameters, Densities, Expansion Coefficients and Perfection of Structure of Cu and of Cu-In α Phase.*

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Spectroscopically pure Cu has a lattice parameter $a_{25} = 3.61491 \text{ \AA}$ (corrected for refraction), and a thermal expansion coefficient $\alpha = 14.87 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ between 15 and 55°C. The measured density d_{25} is $8.9314 \pm 0.0002 \text{ g.cm}^{-3}$ in agreement with the calculated value $d_x = 8.9316$. In the α solid solution region additions of In increase the lattice parameter of Cu according to $a_x = 3.6149 + 0.0091x$ up to $x = 10.4$ ($x = \text{atomic \% In, balance Cu}$). The thermal expansion coefficients between 15 and 65°C of the homogeneous alloys increase from 14.87 (pure Cu) to $17.2 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ at the solid solubility limit (10.4 atomic % In, quenched from 650°C). With the increase of In content the experimental densities become increasingly lower than the calculated ones because of void formation. Upon cold rolling the voids close and the differences disappear. The α phase represents a substitutional solid solution without structural defects. Alloys quenched from the liquid state do not show any microporosity; the voids appear after homogenization at 800°C. Micropore formation is explained by differential shrinkage of the various crystalline fractions formed during solidification, giving rise to internal stresses in the solid alloy. Relief of stresses results in vacancies or micropores, which coalesce into voids upon heat treatment.

Introduction

If point defects are present in crystals in large concentrations, their amounts can be readily determined from lattice parameters and densities. Extra atoms are rarely mentioned, but can be calculated from published lattice parameter and density data, e.g. for the system Si-Ge (Dismukes, Ekstrom & Paff, 1964). Lattice vacancies are reported quite frequently, for instance by Lee & Raynor (1954), Ellwood (1951) and others. Helfrich & Dodd (1962, 1963, 1964) pointed out that the low experimental density obtained might have been caused by microcracks or solidification microporosity. Crawley & Fabian (1966) in their report on Au-Ni alloys arrived at the same conclusion. However, Rider & Roney (1962), confirming the substitutional nature of Pb-In solid solutions, mention that there are discrepancies between experimental and X-ray densities that cannot be due to porosity or structural vacancies.

Thus, it was of interest to know how other homogeneous liquid solutions would behave during crystallization and cooling. For this purpose the system Cu-In was chosen, and the aim of the investigation was: (1) to redetermine the lattice constant of Cu; (2) to check the perfection of the Cu-structure; (3) to redetermine the lattice parameters and room temperature expansion coefficients of the Cu-In solid solutions; (4) to determine structural perfection of the solid solutions; (5) to find an explanation of the microporosity observed.

Experimental approach and materials used

A metal, compound or a solid solution is perfect if the 'actual' number of atoms, or molecules, n' , per unit cell agrees with the number n of occupiable points in the crystallographic sense within the limits of error (Straumanis, 1949, 1953, 1954, 1963). The value of n' is calculated from

$$n' = vdN_0/M \quad (1)$$

where v is the volume of the unit cell, d is the measured density of the homogeneous alloy, having a formula or atomic mass M , and N_0 is Avogadro's number (6.0240×10^{23} unless otherwise stated). In the present case, M was calculated from $\text{Cu}_{(1-x)}\text{In}_x$, taking the weighed amounts of the metals into consideration ($x = \text{atomic \% In}/100$); n' is thus the sum of Cu and In atoms per unit cell.

The perfection of the Cu structure was determined by means of single and polycrystals of 99.999% purity (from ASARCO) and samples of zone-refined, oxygen-free Cu obtained from Professor F. Pawlek (Berlin). Indium (ASARCO) was of the same purity (99.999%). Calculated amounts of Cu and In were weighed to make about 10 g of each of the desired alloys and placed into dry quartz tubes which were evacuated and sealed. The samples were heated to 1150°C. After melting, the two metals were thoroughly mixed by shaking the tube inside or outside the furnace; the tube was then returned to the furnace before the alloy could freeze. This operation was repeated several times. In order to minimize shrinkage voids during solidification and to provide the opportunity for such voids to coalesce to larger ones, the alloys were finally passed

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through the heating zone of the furnace (from the melting point down to 400°C) in about 8 hours. To ensure homogeneity, each ingot was homogenized at 800°C for 14 days and then quenched in water. The weight loss in each sample was less than 1 part in 5000. Finally, filings from both ends of each sample were stress relieved and used for the determination of the lattice constant. Close coincidence of the two values was taken as evidence for sufficient homogeneity.

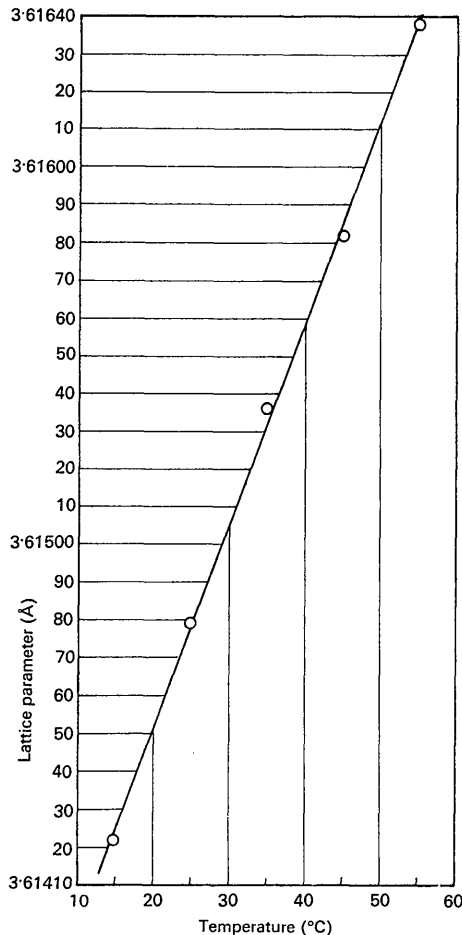


Fig. 1. Lattice constant of Cu versus temperature. Each point is the average of the measurements of 2 films. The probable error (Table 1) is represented by the radius of the circles.

The density of each sample was determined by weighing it in air and xylene at a constant temperature (25.0°C) with a thermostatted semimicro balance. To remove air bubbles adhering on the surface of the samples (in xylene) the beaker was vigorously shaken with a magnetic device under reduced air pressure. The density of each alloy was calculated so as to correspond to that in vacuum.

The lattice constants used for the calculation of v were determined by the asymmetric film method. The filings from the bars were heated in vacuum at the temperature of nearly maximum solubility of In in Cu, 650°C, for 30 minutes and then quenched in cold water. The preparation of the powder mounts is described by Straumanis & Riad (1965). Co $K\alpha$ radiation ($\lambda = 1.78529 \times 1.00202 \text{ \AA}$) produced a distinct $400 \alpha_1\text{-}\alpha_2$ doublet in the high back reflection region. For alloys containing 4 wt.% of In and more, the 420β line showed up clearly enough to be read with the comparator. The lattice parameter for Cu and for each of the alloys was determined from at least 10 films (2 films at each of the 5 temperatures, see Fig. 1). The refraction correction was added to the final values, as shown in Table 1.

Table 1. Lattice parameters a_i of Cu

a_{25} obtained with $\alpha = 14.87 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$

Temperature (°C)	a_i	a_{25}
14.8	3.61422	3.61476
25.0	3.61478	78
35.0	3.61536	82*
45.0	3.61582	75
55.0	3.61638	77
Average		3.61477 ± 0.00002
Refr. corr.		0.00014
		3.61491 \AA

* Excluded.

Lattice parameter and expansion coefficient of Cu

The lattice constants obtained are plotted versus the temperature in Fig. 1. From this straight line the linear thermal expansion coefficient near room temperature, $\alpha = 14.87 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ was calculated. With this value for α , the constants of Cu at the different temperatures were reduced to those at 25°C (Table 1).

Table 2. Experimental density, d , of Cu (X-ray density $d_x = 8.9316 \text{ g.cm}^{-3}$) at 25°C and the actual number of atoms per cell

Cu	Sample	d_{25} (g.cm ⁻³)	n' atoms/unit cell	n''^* atoms/u.c.
ASARCO	Single crystal	8.9332†	4.0007†	4.0002
ASARCO	Polycrystal ann.	8.9334‡	4.0008	4.0003
ASARCO	Polycrystal cold worked	8.9316	4.0000	3.9995
Zone refined	Polycrystal	8.9314‡	3.9999	3.9994

* Using Bearden's (1964, 1967) values $N_0 = 6.2252 \times 10^{23}$ and $A = 1.002056$.

† Propagated error ± 0.0005 (Straumanis, 1963).

‡ Probable error ± 0.0001 to 0.0002 (precision of determination).

Densities and perfection of structure

The final values of the densities of Cu together with the number of atoms per unit cell [equation (1)] are summarized in Table 2. The n' and n'' values show clearly that the element Cu has a perfect structure.

However, the zone-refined Cu (from Germany) with a very low O-content shows, for unknown reasons, a slightly lower density, although both of the samples were spectroscopically pure (only faint traces of Fe were found in the first and of Ag in the German). Slight plastic deformation of the Cu samples did not influence their densities. A 50% reduction of the diameter of the rods by cold working caused a decrease in density by slightly more than the estimated standard deviation. This corresponds to $\sim 5 \times 10^{17}$ vacant sites per cm^3 .

Cu-In solid solutions

Lattice parameters, expansion coefficients and solubility limit

The lattice parameters and expansion coefficients of the alloys were obtained in the same way as shown in Fig. 1 and Table 1 for pure Cu. All the measurements are summarized and compared with the constants of other authors in Fig. 2. It is seen that the maximum solubility of In in Cu at 650°C is 10.4 atomic %, which is about the same as reported by Weibke & Eggers (1934) and by Hume-Rothery, Raynor, Reynolds & Parker (1940). These authors gave 10.4 and 10.35 at. % respectively. Owen & Roberts (1952) and Jones & Owen (1953) found ~ 10.5 at. %.

The expansion of the Cu lattice due to dissolution of In is in accord with Vegard's straight line:

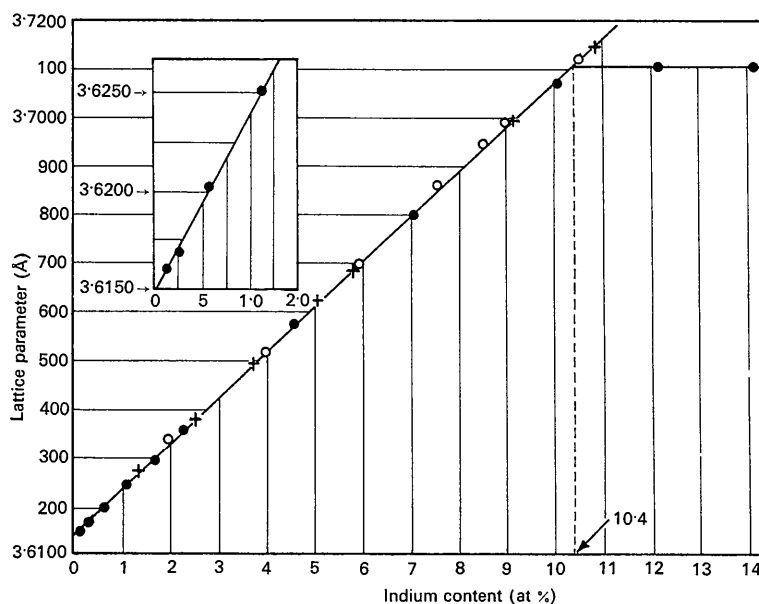


Fig. 2. Lattice parameters of the α -phase of Cu-In solid solutions at 25°C (full circles). Inset: a versus small amounts of In in Cu. All powder samples quenched from 650°C . +, Owen & Roberts; O, Jones & Owen.

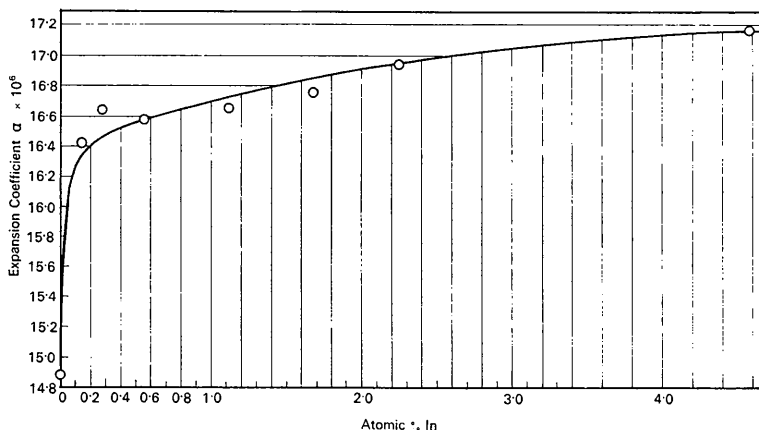


Fig. 3. Room temperature thermal expansion coefficients (between 15 and 65°C) of Cu and of Cu-In alloys versus In concentration.

$$a_x = 3.6149 + 0.0091x, \quad (2)$$

where a_x is the lattice parameter of the solid solution at any concentration, x , up to 10.4 at.% of In in Cu. The average deviation of the experimental values from the calculated ones [equation (2)] is within $\pm 0.0003 \text{ \AA}$. The a constants of 2 alloys of the saturated α -phase (containing 12.15 and 14.18 at.% In) were 3.7104 and 3.7105 \AA respectively at 25°C.

The thermal expansion coefficients are plotted *versus* the In content in Fig. 3; they increase markedly at low In concentrations with the In content.

Densities and imperfections in the alloys

While the reproducibility of the measured density of pure Cu was good (Table 2), there were great difficulties in reproducing the densities of the solid solutions. Remelted and slowly cooled alloys always showed large

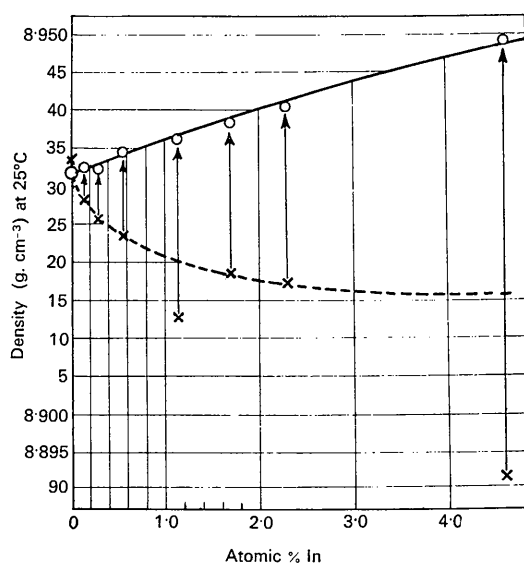


Fig. 4. Densities of Cu-In solid solutions within the range of the α -phase. x, maximum densities of the slowly cooled alloys (dashed curve); O, densities of rolled samples (70% reduction); solid curve, theoretical densities.

differences in d_{25} , although the reproducibility of the density measurements without remelting or other treatment was excellent. The maximum density obtained for each solid solution is shown in Fig. 4 (broken line). Two of the density points are far below the curve. These alloys were remelted only twice. Microscopic examination of sectioned samples revealed even at low magnification spherical and elongated voids. The number of these increased with increasing concentration of In.

Since the Cu-In alloys are soft, the voids collapsed under application of pressure. A 70% reduction in the diameter of the alloy rods by cold rolling proved to be sufficient to eliminate almost all the voids. The density increased considerably. For stress relief the samples were annealed at 550°C, which resulted in an additional slight increase in density approaching the calculated value (Table 3).

As can be seen from Table 3, compression completely closed the voids. Measured and calculated densities were nearly identical in all cases and the number of formula units, n' , came out to be equal to 4.0000 within the experimental error. Since the alloy containing 10.18 at.% In (Fig. 2) was too close to the solubility limit of In in Cu, its density was not determined.

The factors causing void formation

As is evidenced by Fig. 4 and Table 3, the total volume of voids increases with increasing In content, possibly reaching a maximum at the phase boundary. Since low rate of freezing did not eliminate these defects and since the temperature interval of solidification increases with increasing solute concentration (see Fig. 5), it is likely that the extent of void formation (microporosity) is related to the degree of shrinkage. Such a possibility was suggested by Helfrich & Dodd (1962, 1963, 1964). That In introduction is the cause of the defects is further evidenced by the absence of voids in pure Cu, which freezes at a single temperature.

To investigate further the mechanism of void formation several Cu-In alloys were quenched from the liquid state and examined prior to annealing. No micropores could be detected in these, even at 750 times

Table 3. Densities of primary Cu-In solid solutions at 25°C, X-ray densities (25°C) and number of formula masses, n' , per unit cell of the rolled and recrystallized samples

Indium		Formula mass	Densities in g.cm ⁻³			n' heating
Weight %	Atomic %		Before rolling	Rolling and heating	d_r	
—	0.000	63.54	8.9334	8.9316	8.9316	—
0.25	0.138	63.611	8.9284	—	8.9320	—
0.50	0.277	63.682	8.9258	8.9322	8.9331	3.9996
1.00	0.556	63.825	8.9238	8.9344	8.9339	4.0002
2.00	1.117	64.112	8.9123	8.9364	8.9363	4.0001
3.00	1.683	64.403	8.9187	8.8382	8.9390	3.9996
4.00	2.250	64.696	8.9175	8.9404	8.9412	3.9997
8.00	4.587	65.892	8.8920	8.9492	8.9490	4.0003

Precision of density determinations: $\pm 0.0004 \text{ g.cm}^{-3}$ or better.

Propagated error of n' : ± 0.0005 .

magnification [Fig. 6(a) and (b)], but the measured densities were still considerably lower than those calculated on the basis of the unit-cell sizes. After subsequent annealing between 700 and 800°C for five days, pores became clearly visible and the number of pores increased with increasing In content (Fig. 7). The alloys were now, of course, homogeneous. The voids resembled those obtained by slow cooling of the samples of Fig. 4. The processes of void formation have been discussed by Clarebrough (1966), Clarebrough, Humble & Loretto (1967), Ruedl, Delavignette & Amelinckx (1962), Barrett & Massalski (1966), and Ovsienko (1968). It is believed that initially vacancies or sub-microscopic pores appear during solidification and that these coalesce to larger voids upon subsequent annealing or during very slow cooling. Vacancies that are close to the surface may migrate outward and disappear.

A discussion on void formation is also given by Maciolek, Mullendore & Dodd (1967) but no satisfactory mechanism is described. The process of void formation and collapsing within solid solutions is very similar to that reported by Doyle, Hulm, Jones, Miller & Taylor (1968), who removed, partially or completely, the 15.5% vacancies present in TiO (Straumanis & Li, 1960) by application of high pressure and temperature (1650°C).

A mechanism of void formation that is proposed here and does not seem to have been mentioned in the literature is *differential shrinkage*. This phenomenon is explained conveniently with the aid of the phase diagram shown in Fig. 5. At a given composition of the liquid, say x_1 , above the α phase (Fig. 5, arrow), an alloy of composition x_2 rich in Cu crystallizes first. Since the liquid now becomes richer in indium, the solidification temperature decreases, and crystals continually richer in indium precipitate. The last traces of the liquid finally freeze at point x_1 . Thus the alloy contains crystallites in which the amount of indium has increased during cooling from x_2 to x_1 . Because of the considerable difference in the expansion coefficients (Fig. 3), the Cu-rich fractions will shrink less than the surrounding In-rich fractions, causing strains and stresses to be built up. These are relieved through formation of vacancies, the total volume of which will correspond to the difference in shrinkage of the various fractions. The voids are formed subsequently through coalescence as was described above.

As the process of crystallization and cooling of a solid solution is complicated, only very approximate calculations can be made based on the Cu-In equilibrium diagram (Hansen, 1958), on the lattice parameters of the respective crystalline fractions and on their room temperature expansion coefficients; the high temperature coefficients are unknown. For convenience, single unit cells of each of the alloys of the composition x_1 and x_2 (Fig. 5) were considered, expanding from 25°C to the temperature of total solidification t_1 , and disregarding the diffusion of Cu and

In atoms. The increase in volume, ΔV_1 , of the cell of the composition x_1 is then

$$\Delta V_1 = 3\alpha_1 a_1^3 (t_1 - 25) \quad (3)$$

and that of the cell x_2 is

$$\Delta V_2 = 3\alpha_2 a_2^3 (t_1 - 25), \quad (4)$$

where $3\alpha_1$ and $3\alpha_2$ are the volume expansion coefficients (read from Fig. 3), a_1 and a_2 are the lattice parameters of the alloys of the composition x_1 and x_2 , which is read from the enlarged Cu-In equilibrium diagram of the α -phase (Hansen). Then a_1 and a_2 can be calculated from equation (2). The difference $\Delta V_1 - \Delta V_2$ gives the total volume of the vacant sites V_v for the two unit cells, in \AA^3 :

$$V_v = 3(t_1 - 25) (\alpha_1 a_1^3 - \alpha_2 a_2^3). \quad (5)$$

The same V_v will also be obtained if the solid solution is cooled from t_1° to 25°C.

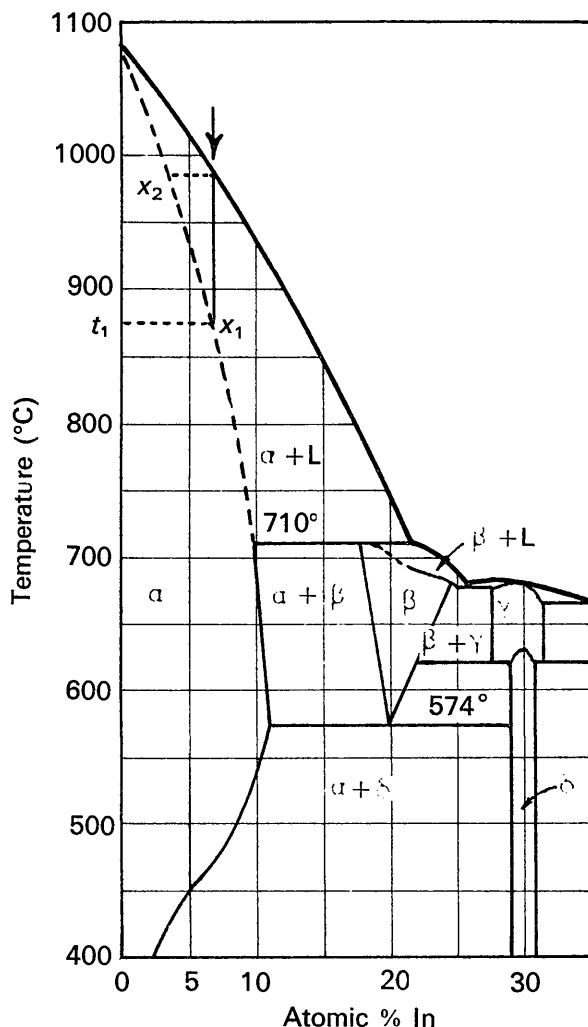


Fig. 5. The α -phase of the Cu-In system (Hansen).

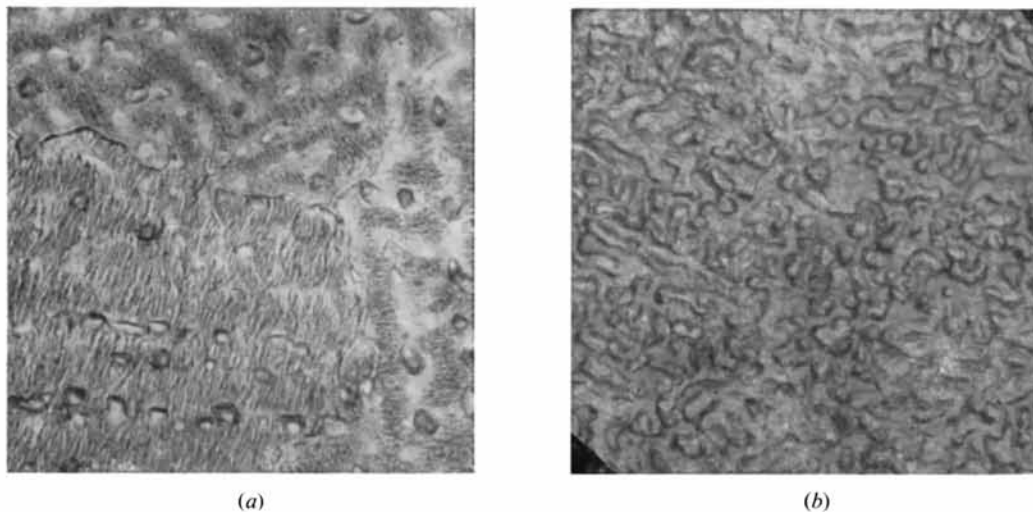


Fig. 6. Sections through Cu-In alloys quenched from the liquid state ($\sim 100^\circ$ above m. p.): (a) 2.25 at.% In; (b) 10.18 at.% In. Absence of microporosity. The dots represent the In-rich phase, severely attacked by the etchant, $\times 750$.

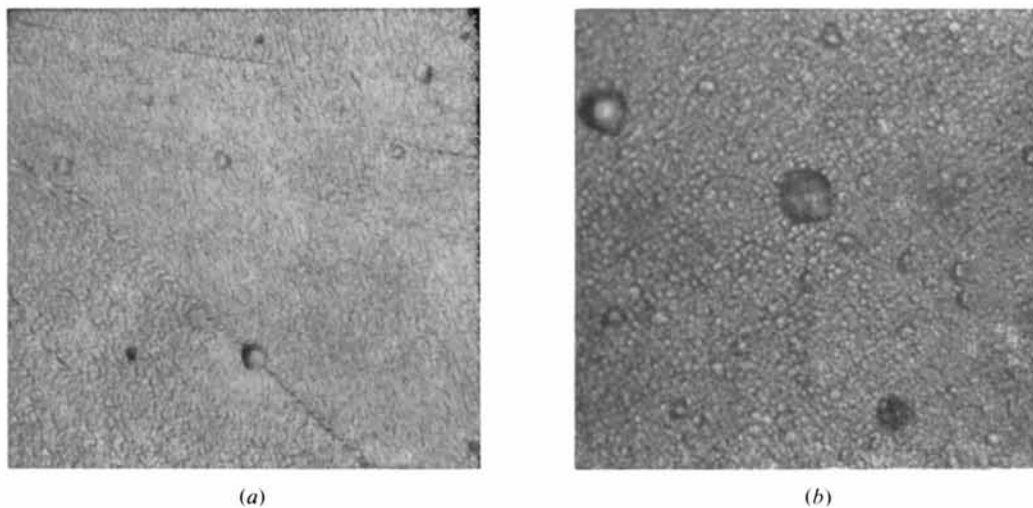


Fig. 7. Voids formed due to coalescence of vacancies during heat treatment. Alloy (a) and alloy (b) (Fig. 6), $\times 750$.

The volume V_v can also be expressed in terms of single vacant site concentration, c (per unit cell) and per In atom, since the two cells contain $8 \text{ Cu}_{1-x}\text{In}_x$ formula weights, and the x of the precipitates at the beginning and end of crystallization, x_1 and x_2 , are known (Table 3):

$$c = 8V_v/(a_1^3 + a_2^3)(x_1 + x_2). \quad (6)$$

The results of the calculation are summarized in Fig. 8 (curve a). The second curve, b , was obtained by computing n' from equation (1), in which for d the maximum densities prior to rolling, and for M the respective formula masses (Table 3) were substituted. The number of single vacancies, c (per unit cell and In atom) is then:

$$c = (4 - n')/n'x \quad (7)$$

where $n'x$ is the number of In atoms per unit cell at the respective composition (Straumanis, 1949, 1953, 1954).

The agreement between the two curves of Fig. 8 is not very good (curve b is logarithmic), however, the deviations are understandable if the very approximate conditions of calculation of curve a are considered; as no diffusion was assumed, naturally, the first Cu-rich fractions, where the difference in α_1 and α_2 is largest, should produce a larger calculated total vacancy space than is actually obtained. That with increasing In concentration more vacancies were observed than were calculated from the expansion coefficients α_1 and α_2 may be due to coring (Rhines, 1956), whereby the β phase (Fig. 5) is also segregated, probably having expansion coefficients substantially different from α_1 and α_2 . It may also be that more vacancies are produced than follows from equations (5) and (6), probably due to diffusion of In into Cu-rich fractions (Kirchendall effect, see e.g. Hauffe, 1966).

Differential shrinkage of crystalline fractions seems to be an important factor in producing vacancies and voids during cooling of solid solutions.

Conclusions

The formation of vacancies and voids during the cooling of the solid solutions from the liquid state is not an equilibrium process, but results from annihilation of internal stresses. This is evidenced by the fact that solid solutions without defects can be prepared by vapor-diffusion [see for instance Al-Zn (Helfrich & Dodd, 1962)].

Samples within the solid solubility range in which the components have nearly the same expansion coefficients (α) should not produce significant amounts of vacancies or voids during cooling and subsequent heat treatment.

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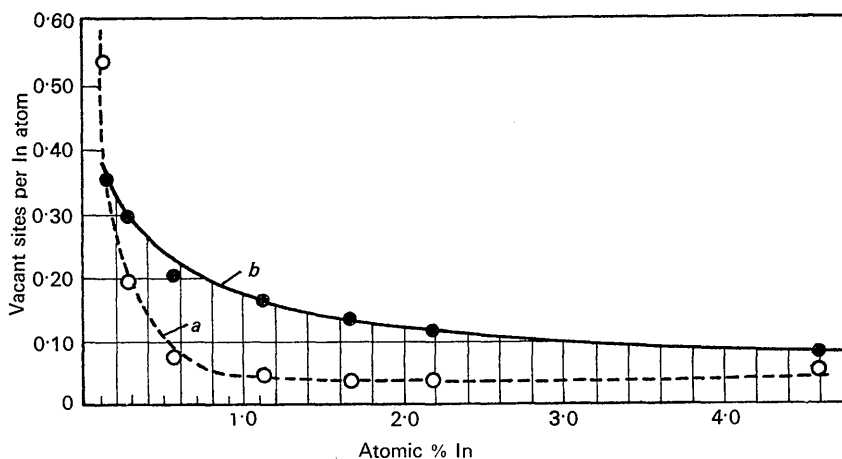


Fig. 8. Vacant sites per In atom versus In concentration of Cu-In solid solutions. a , calculated from phase diagram and α of Fig. 3; b , from lattice parameters, composition and the maximum of experimental densities (Fig. 4).

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X-ray Diffraction from a Crystal Containing Isolated Imperfections

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The X-ray scattering amplitude of a crystal containing isolated imperfections is expressed in terms of the Fourier transform of the atomic displacement vectors. The amplitude contains only the local properties of the imperfections which are 'seen' by the incident beam of a finite size. Disruption of the Borrmann transmitted (or diffracted) beam, narrowing of the dynamical diffraction range and 'diffuse' scattering caused by dynamically diffracted X-rays are some of the results obtained from the calculated amplitude. Black, white and black-white images in topographs are explained by the present theory. Image contrast is also discussed in terms of the thickness of the crystal.

1. Introduction

Recent interest in X-ray diffraction from a single crystal has been centred on the fine structure in a Laue spot. This fine structure is closely related to imperfections in crystals. The techniques of X-ray topography 'magnify' at high resolution a Laue spot from a large portion of a crystal.

In the X-ray topographs obtained from an imperfect crystal, one observes black and white images superimposed on a background. For a *thin* crystal where the product of the linear absorption coefficient μ and the thickness L is less than 1, defects may appear as black (stronger intensity) images accompanied by a faint anomalous transmission (or diffraction) beam trace. For *intermediate* thickness where $1 < \mu L < 10$, the topographs show primarily white images. Black

images also appear as well as black-white contrast images. Usually one should expect complicated patterns for these cases. For a *thick* crystal ($\mu L > 10$), the Borrmann (anomalous transmission) effect (Borrmann, 1941, 1950) becomes dominant and is accompanied by white images of good contrast. Black images or non-uniform intensity distribution may still be observed in the topographs. The above qualitative description of images is based on casual observation of topographs. More complicated patterns including interference effects, etc. are also observed.

The primary objective of this paper is to present some of the explanations for the formation of the images mentioned above, by use of a previously formulated general theory of scattering from an imperfect crystal (Kuriyama, 1967*a*, 1968*b*). This theory has been derived with a large degree of rigor. Wherever approx-