

Isothermal coarsening of liquid inclusions in aluminium-copper alloys

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An isothermal coarsening model was introduced for liquid inclusions in homogeneous aluminium–copper alloys. Discrepancies between experimentally measured and analytically predicted variations of the solid–liquid interface area per unit volume of liquid, Sv , the average inclusion diameter, \bar{D} , and the number of inclusions per unit volume of solid, Nv , with isothermal coarsening time were attributed to experimental evaluation errors, to the inaccuracy of some assumptions made or values adopted for certain system variables and to the effect of melting and solidification kinetics. At constant fraction liquid, isothermal coarsening slows down with increasing copper concentration.

1. Introduction

During certain metallurgical operations, such as high temperature metal forming, annealing or homogenization, partial remelting of the solid may sometimes occur, in the form of liquid inclusions. In a fairly homogeneous alloy liquid will appear during reheating both at the grain boundaries, and within the grains in the form of randomly situated liquid inclusions. The remelting pattern depends primarily on the heating rate, as reported elsewhere [1]. If the solid–liquid interfacial energy is isotropic, intragranular inclusions are usually spherical and coarsen with time during the heat-treatment.

The isothermal coarsening of dendritic solid surrounded by liquid was studied previously [2, 3] and coarsening kinetics were analytically established and compared with experimental results [4]. This analysis was extended to the isothermal coarsening of spherical particles surrounded by liquid [5]. The average particle size increased with isothermal holding time, whereas the number of particles per unit volume and the total solid–liquid interface area per unit volume of solid decreased.

An analogous study is reported here on the isothermal coarsening behaviour of liquid inclusions in the aluminium–copper alloy system. The validity of an analytically derived expression

for coarsening kinetics was tested with alloys of various copper contents, which were isothermally held at temperatures corresponding to the same fraction liquid for all alloys.

2. Experimental procedure

Alloys of aluminium–2, 4 and 6 wt % copper were prepared and air-cast in a copper-chill mould in the form of 6.5 mm diameter rods, which were fully homogenized by a treatment at 773 K for several days. The homogenized rods were subsequently sectioned into segments, which were reheated (each in contact with a thermocouple bead) in a resistance furnace at various temperatures. These annealing or coarsening temperatures were selected from the equilibrium phase diagram so that 10%, by weight, liquid was present in each alloy and were controlled within ± 1 K. In each case the temperature was reached at a heating rate of about 0.033 Ksec^{-1} . The necessity for a uniform heating rate was emphasized by Lalor elsewhere [1]. Samples were pulled out of the furnace and quenched in water after 0.08, 1, 10, 50 and 100 h isothermal holding. They were subsequently sectioned, polished, etched and studied by quantitative metallography. In each sample the liquid–solid interface area per unit volume of liquid inclusion, the number of inclusions per unit volume of solid matrix, and the average inclusion

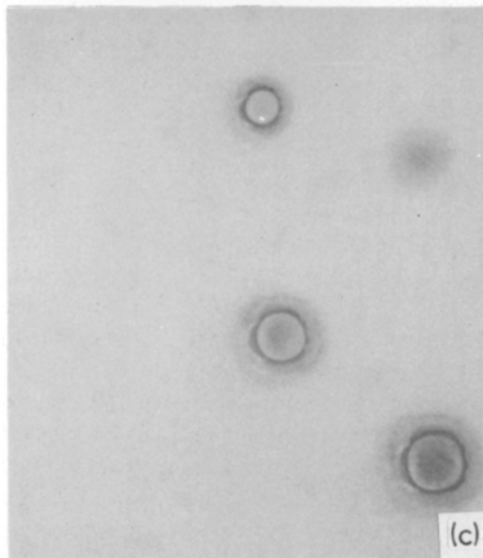
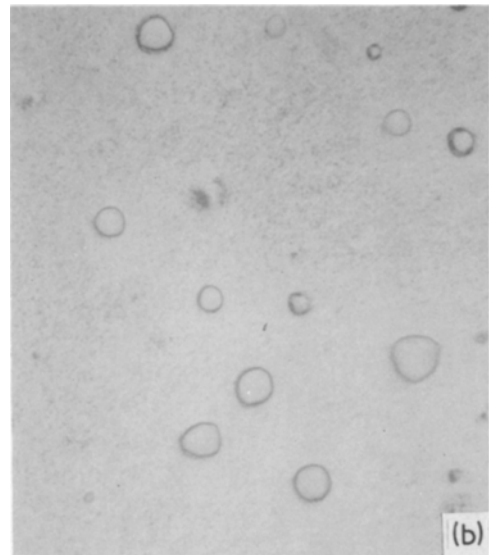
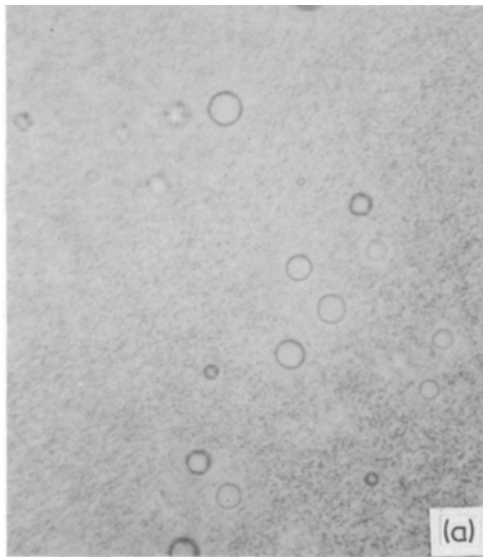


Figure 1 Photomicrographs of Al-4 wt% Cu alloy specimens held isothermally at 881 K for (a) 1 h, (b) 10 h and (c) 100 h, $\times 800$.

Results from the quantitative evaluation of inclusion geometry are given in Fig. 3 to 5. Fig. 3 illustrates the variation of Sv/Sv_0 with coarsening time, Fig. 4 illustrates that of \bar{D}/\bar{D}_0 and Fig. 5 that of Nv/Nv_0 , where Sv is the solid-liquid interface area per unit volume of liquid, \bar{D} is the average liquid inclusion diameter, Nv is the number of inclusions per unit volume of solid, and Sv_0 , D_0 and Nv_0 are values of these respective parameters taken at time 0. These data, replotted on logarithmic axes, led to the following expressions:

$$\frac{Sv}{Sv_0} = 0.83 C_0^{0.32} t^{-0.30} \quad (1)$$

$$\frac{\bar{D}}{\bar{D}_0} = 1.41 C_0^{-0.53} t^{0.30} \quad (2)$$

$$\frac{Nv}{Nv_0} = 0.54 C_0^{0.40} t^{-0.35} \quad (3)$$

diameter, were evaluated using techniques described by Underwood [6] for spheroid inclusions of nonuniform size and applied previously [5].

3. Results and discussion

The variation in inclusion geometry with coarsening time for aluminium-4 wt% copper is illustrated in Fig. 1. The dependence of inclusion size on alloy composition is illustrated in Fig. 2 for aluminium-2, 4 and 6 wt% copper alloy specimen, isothermally coarsened for 100 h at 909, 881 and 858 K, respectively. These temperatures correspond to a weight fraction liquid of 0.1 for all three alloys. Under these operating conditions liquid inclusions coarsen faster in the more dilute alloy.

where C_0 is the wt% copper in the alloy and t is isothermal coarsening time in h. Coarsening is assumed to be conducted at a temperature at which 10% by weight of liquid is present.

3.1. Model for isothermal coarsening of liquid inclusion

The interaction between two liquid inclusions will first be considered. Results will subsequently be

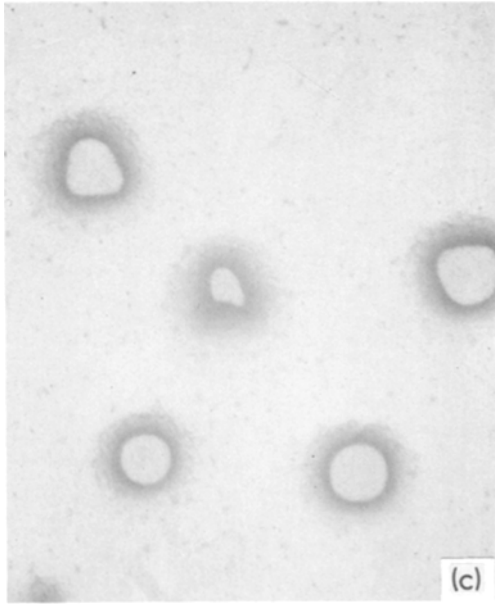
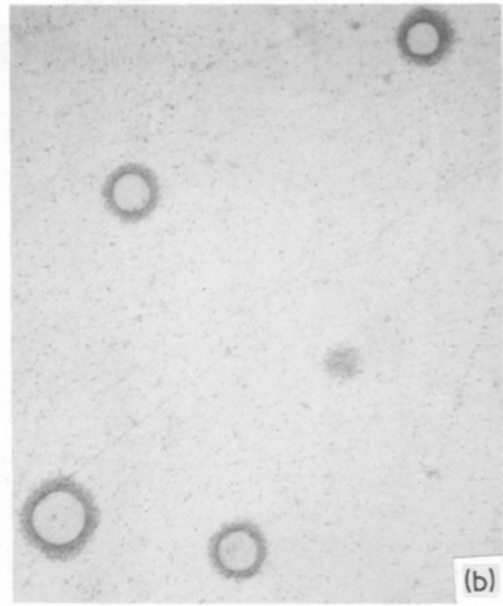
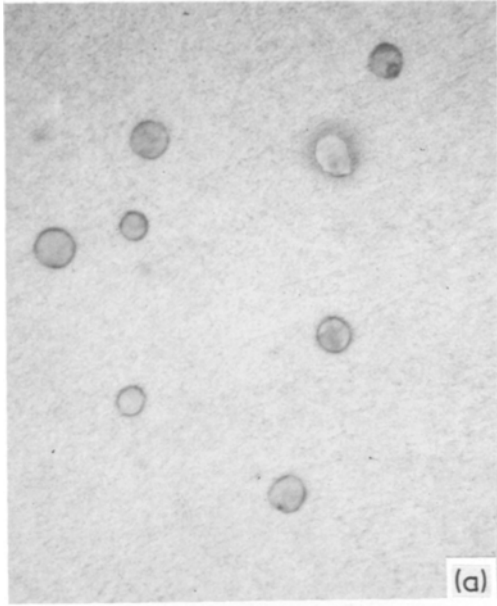


Figure 2 Photomicrographs of Al-Cu specimens held isothermally for 100 h at various temperatures, $\times 800$. Compositions and temperatures are (a) 2 wt % Cu, 909 K, (b) 4 wt % Cu, 881 K and (c) 6 wt % Cu, 858 K.

where C_L^a is the concentration of solute in the liquid at temperature T in equilibrium with a surface of radius a (wt %), C_s is the concentration of solute in the solid at distance r and temperature T (wt %), r is the distance from the centre of the inclusion (m), k is the equilibrium partition ratio, ρ is the density (kg m^{-3}), a is the radius of smaller inclusion (m), t is the time (sec), D is the diffusivity of solute in the solid ($\text{m}^2 \text{sec}^{-1}$). Equation 4 may be written as:

$$C_s^a \left(\frac{1-k}{k} \right) \rho 4\pi a^2 \left(\frac{da}{dt} \right) = D \rho 4\pi r^2 \frac{dC_s}{dr} \quad (5)$$

where C_s^a is concentration of solute in the solid at temperature T in equilibrium with a surface of radius a (wt %).

Assuming $C_s^a \approx C_s$, rearranging and integrating between $r = a$ and $r = \infty$:

$$\int_0^\infty \frac{C_s(1-k)a^2}{kDr^2} \left(\frac{da}{dt} \right) dr = \int_{C_s^a}^{C_s^\infty} dC_s. \quad (6)$$

Hence,

$$\frac{da}{dt} = \frac{Dk}{C_s(1-k)a} (C_s^\infty - C_s^a). \quad (7)$$

Assuming, as previously [4], that:

generalized by considering a system of n inclusions, and formulating an equation which describes the radii of all inclusions at any time and can be solved by means of a digital computer.

Assume an isothermal system consisting of two spherical inclusions of different radii in homogenized aluminium-rich α -phase.

Following an analysis analogous to that published previously [5], the solute balance for a shrinking liquid inclusion at temperature T can be written as:

$$C_L^a (1-k) \rho 4\pi a^2 \left(\frac{da}{dt} \right) = D \rho 4\pi r^2 \frac{dC_s}{dr} \quad (4)$$

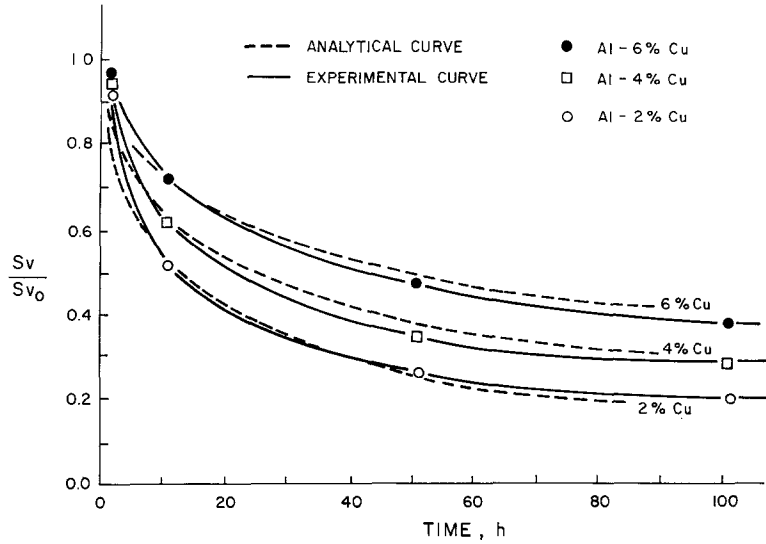


Figure 3 Variation of Sv/Sv_0 with coarsening time for Al-2, 4 and 6 wt% Cu alloy specimens. Comparison of experimental and analytical curves.

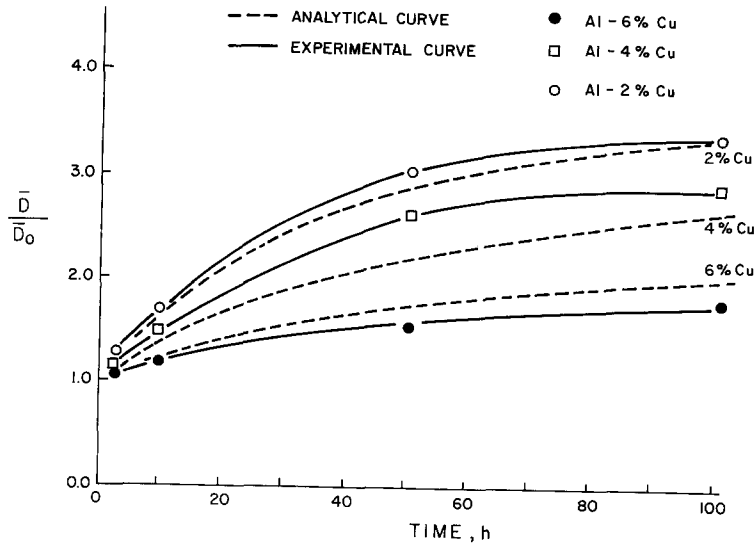


Figure 4 Variation of \bar{D}/\bar{D}_0 with coarsening time for Al-2, 4 and 6 wt% Cu alloy specimens. Comparison of experimental and analytical curves.

$$C_s^\infty - C_s^a \approx C_s^R - C_s^a \quad (8) \quad (K/\%), \text{ and } \sigma \text{ is solid-liquid interface energy (J m}^{-2}\text{). Therefore,}$$

it follows [4] that :

$$C_s^\infty - C_s^a \approx \frac{2\sigma T}{m_s H} \left[\frac{1}{R} - \frac{1}{a} \right] \quad (9) \quad \frac{da}{dt} = \frac{2\sigma D T k}{C_s(1-k)m_s H a} \left[\frac{1}{R} - \frac{1}{a} \right]. \quad (10)$$

where C_s^R is the concentration of solute in the solid at temperature T in equilibrium with a surface of radius R (wt%), R is the radius of the larger inclusion (m), H is the volumetric heat of fusion ($J m^{-3}$), m_s is the slope of the solidus

Equation 10 gives the rate of disappearance of the smaller inclusion.

Applying the previous analysis [5] to a system of $n > 2$ spherical inclusions and assuming that the total volume of liquid remains constant during isothermal holding, it can be established that:

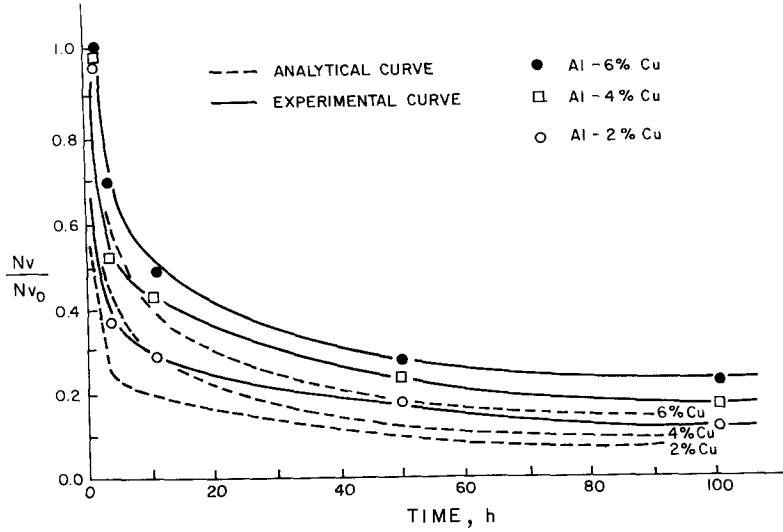


Figure 5 Variation of Nv/Nv_0 with coarsening time for Al-2, 4 and 6 wt %Cu alloy specimens. Comparison of experimental and analytical curves.

$$\frac{dr_i}{dt} = \sum_{j=1, j \neq i}^n \left\{ \frac{-2\sigma D T k}{C_s(1-k)m_s H r_i} \left(\frac{1}{r_i} - \frac{1}{r_j} \right) \right\}$$

$$\mathcal{H}(r_j - r_i) + \frac{-2\sigma D T k r_j}{C_s(1-k)m_s H r_i^2} \left(\frac{1}{r_i} - \frac{1}{r_j} \right) \mathcal{H}(r_i - r_j)$$

with

$$\mathcal{H}(\alpha - \beta) = 0, \alpha < \beta$$

$$\mathcal{H}(\alpha - \beta) = 1, \alpha > \beta$$

where r_i and r_j are the radii of any two inclusions i and j . The radius of a spherical inclusion i as a function of time is then:

$$r_i = r_0 + \int_0^t \left(\frac{dr_i}{dt} \right) dt. \quad (12)$$

The surface-to-volume ratio at any time t is given by:

$$Sv = \frac{\sum_{i=1}^n 4\pi r_i^2}{\sum_{i=1}^n \frac{4}{3}\pi r_i^3}, \quad (13)$$

thus, the radii of all the inclusions of the system can be calculated at any time by evaluating Equations 11 and 12 by means of a digital computer. The system can therefore, be described at any time. Sv may be calculated from Equation 13.

At a given time t after initiation of coarsening:

$$Sv = Sv_0 + \Delta Sv. \quad (14)$$

The following values were adopted [2] for the system variables in Equation 11: $m_s = 19.82$ K/% Cu, $k = 0.17$, $\sigma = 5.02 \times 10^{-2} \text{ J m}^{-2}$ (no information is available on the composition dependence of σ), $H = -1.15 \times 10^9 \text{ J m}^{-3}$. For Al-2 wt % Cu, $D = 9.4 \times 10^{-14} \text{ m}^2 \text{ sec}^{-1}$ [7], $C_s = 1.2$ 2t % Cu and $T = 909$ K. For Al-4 wt % Cu, $D = 5.0 \times 10^{-14} \text{ m}^2 \text{ sec}^{-1}$ [7], $C_s = 2.6$ wt % Cu, $T = 881$ K, and for Al-6 wt % Cu, $D = 2.0 \times 10^{-14} \text{ m}^2 \text{ sec}^{-1}$ [7], $C_s = 3.8$ wt % Cu and $T = 855$ K.

The computer program assumed an initial population of 19 spheres, selected randomly from specimens which were heated for a very short time (300 sec) and were subsequently quenched. The program printed out the radius of each spherical inclusion and the Sv for each specimen, at any given time. The disappearance (solidification) of a liquid inclusion or the annihilation of its radius after considerable coarsening time shows up as a decrease in Nv . In computing the average inclusion diameter, \bar{D} , inclusions of zero radii were not considered.

The analytical curves of coarsening kinetics are plotted in Fig. 3 to 5, together with the experimental curves, for comparison purposes. The observed discrepancies could be attributed to: (a) experimental errors in quantitative metallo-

graphic evaluations; (b) errors in the estimation of diffusivities and interfacial energy; (c) inaccuracy of the assumption that heat of fusion and solid-liquid interfacial energy are independent of composition and; (d) effect of melting and solidification kinetics.

4. Conclusions

(1) The isothermal coarsening model introduced herein predicts approximately the coarsening kinetics of liquid inclusions in homogeneous aluminium-2, 4 and 6 wt % copper alloys.

(2) At a given volume fraction liquid the more dilute alloys coarsen faster than do the more concentrated alloys.

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