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Phenomena and microstructure at the aluminium–lead interface

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An attempt has been made to study the precipitation behaviour of an Al–Pb alloy with respect to the unavoidable sedimented lead layer. The effect on the melt of Lorentz force on which is superimposed an external current is discussed briefly. This method could be further sophisticated to generate a higher volume fraction of heavier component in the matrix, by suitably adjusting the process parameters which influence the convection, and also by varying the applied external current density.*

1. Introduction

Alloy systems like aluminium–lead, where a pronounced miscibility gap exists, pose a great challenge to material scientists. The separation of such liquids is generally explained by considering the thermodynamics of non-ideal solutions. If the attraction of Al atoms to Pb atoms in the Al–Pb system is greater than that of Al–Al or Pb–Pb, the enthalpy of mixing is negative. If on the contrary the forces of attraction are greater between like Al–Al and Pb–Pb atoms, a clustering tendency is expected, thereby resulting in unmixing of the liquid to form two phases [1]. Both the elements aluminium and lead are almost insoluble in each other in liquid (up to about 1273 K) as well as in solid states. The monotectic point lies at 0.19 at % Pb and the second liquid L_2 dissolves up to about 2 at % Al. The solubility of lead in aluminium at 1103 K is about 2 at % and that of aluminium in lead is still less [2]. Because of the wide freezing range and a large difference in densities, a gravity sedimentation of lead in conventional solidification techniques is inevitable. Some attempts have been made in recent years to disperse lead homogeneously into the aluminium matrix through standard casting methods [3–5].

The object of this present work is to shed some light on the phenomena occurring in the Al–Pb system (Fig. 1) [6] with a particular reference to the morphology of the microstructure at the interface of the sedimented lead layer and aluminium.

2. Experimental procedure

Commercial aluminium in cylindrical form and lead of high purity, corresponding to an alloy of composition AlPb20, were melted in an alumina crucible placed vertically and centred in an induction coil operating at high frequencies. After initially raising the temperature up to 1393 K (which corresponds to the homogenizing temperature of an alloy of the above composition), Ti was added in the form of a commercial master alloy AlTi5B0.6. At the above temperature, an external current of a certain density (about

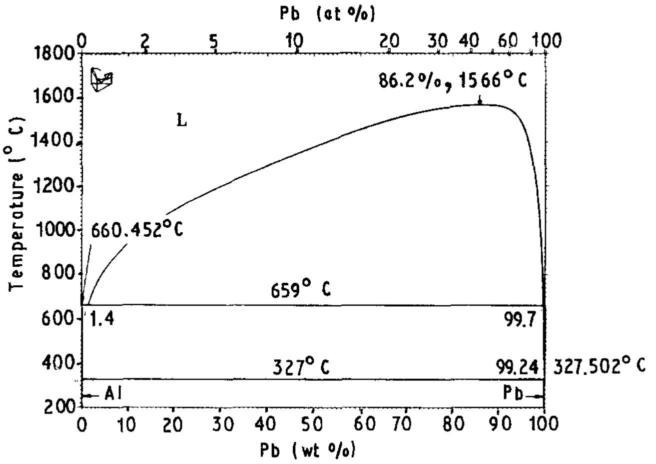

Figure 1 Al–Pb binary phase diagram [6].

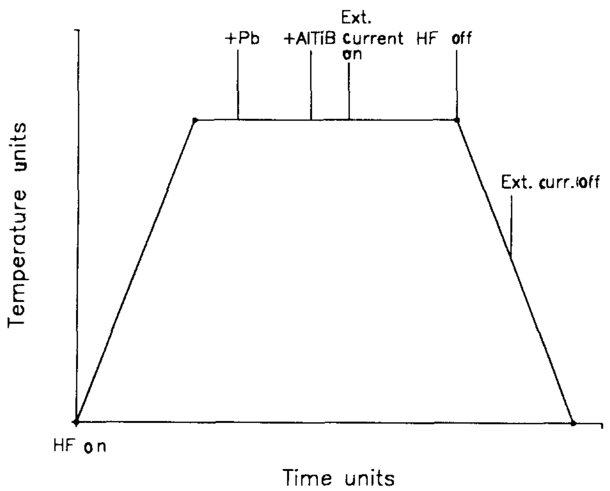

Figure 2 Schematic representation of an experimental cycle.

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*This paper is dedicated to Prof. Dr.-Ing H. Wever, T U Berlin on his 70th birthday.

60 μA cm^{-2}) was applied to the system (Fig. 2). The melt was then allowed to cool in the crucible, the applied current being held constant until the entire melt had solidified. The solidified specimen was 6 cm in length and 1.5 cm in diameter. A separate experiment was conducted with the same induction melting equipment, this time to register only the temperature at the Al–Pb interface during the solidification with the help of an NiCr–Ni thermocouple of 1 mm diameter which was sheathed in a thin alumina tube.

To facilitate the microscopic investigation, a longitudinal and a cross-sectional specimen were cut out of the solidified sample.

3. Theoretical considerations

An Al–Pb alloy of composition between the limits of the miscibility gap when heated to a temperature above the "dome" exists as a homogeneous single-phase liquid. Under gravity conditions on cooling beyond the liquid phase boundary it transforms itself into two liquids, one rich in aluminium and the other in lead, thereby lowering the free energy of the system. The two liquids differ in their densities, thus resulting in preferential sedimentation of the heavier liquid. On cooling down to the monotectic temperature and below, there is a precipitation of solid aluminium from the Al-rich liquid and a liquid phase rich in lead. Some more Al crystallizes on the already existent Al phase on further cooling. Finally at the temperature of the freezing point of lead the entire alloy solidifies.

In order to stabilize the lead dispersions in the Al matrix, a method has to be adapted to slow down, if not completely overcome, the downward Stokes motion of the Pb droplets formed. One could envisage methods like rapid solidification or ultrasonic treatment of the melt coupled with rapid solidification or solidification under reduced gravity [7].

4. Results and discussion

In this work a high-frequency alternating current is passed through a coil surrounding the cylindrical charge. The changing magnetic field induces eddy currents in the conducting charge, generating Joule heat which heats up the material from the surface and is transmitted radially towards the centre through conduction, leading to subsequent melting of the charge. The eddy currents interact also with the magnetic field to produce Lorentz forces which act on the melt. To understand precisely the nature and magnitude of current and field, their distribution and the ensuing resultant body forces affecting the melt, Maxwell's electromagnetohydrodynamical (EMH) equations together with the Navier–Stokes equations [8, 9] have to be solved, taking into consideration the differences in electrical conductivities, surface tensions, viscosities and specific densities of aluminium, lead, Al_3Ti and TiB_2.

Because of the wide difference in specific densities between Al and Pb (2.35 and 10.3×10^3 kg m^{-3}, respectively) at 1273 K the fluid forces in the melt tend to be asymmetrical, giving rise to a convective motion–turbulent, if the Reynolds number Re is greater than a certain critical value Re_{cr}, and laminar if $Re \ll Re_{cr}$. Superimposition of direct current applied externally to the system complicates the nature of the generated body force fields and their distribution. To compute these factors is not the aim of this work.

Fig. 3a shows a longitudinal section of the solidified specimen without treatment. Fig. 3b shows a longitudinal section of the specimen solidified under applied external current. On the aluminium side, in the immediate neighbourhood of the Al–Pb boundary, spheroids (some of them oblate) of lead of various sizes, ranging approx. from 5 to 60 μm in diameter, were observed (Fig. 3b).

If we suppose that the body forces emanating from the EMH effects set a thin layer of fluid lead column in motion, then the initial cylindrical form of the lead layer can collapse into a row of spheroidal dispersions which are partly oblate [10, 11]. Their near-spherical shape points to a low anisotropy of Al–Pb interfacial energy. On cooling the melt in the crucible, those dispersions which were larger than 25 μm migrate downwards with the Stokes' velocity [12], which is given by the equation

$$V_s = 2r^2a(\rho_{Pb} - \rho_{Al})/9\eta$$

where ρ_{Pb}, ρ_{Al} are the densities of lead and aluminium, respectively, r = radius of the dispersoid, η = viscosity of lead and a = acceleration due to gravity. The growth of the spheres is either the result of a coalescence due to collision of the smaller dispersions or due to Ostwald ripening.

The other effect which could possibly explain the

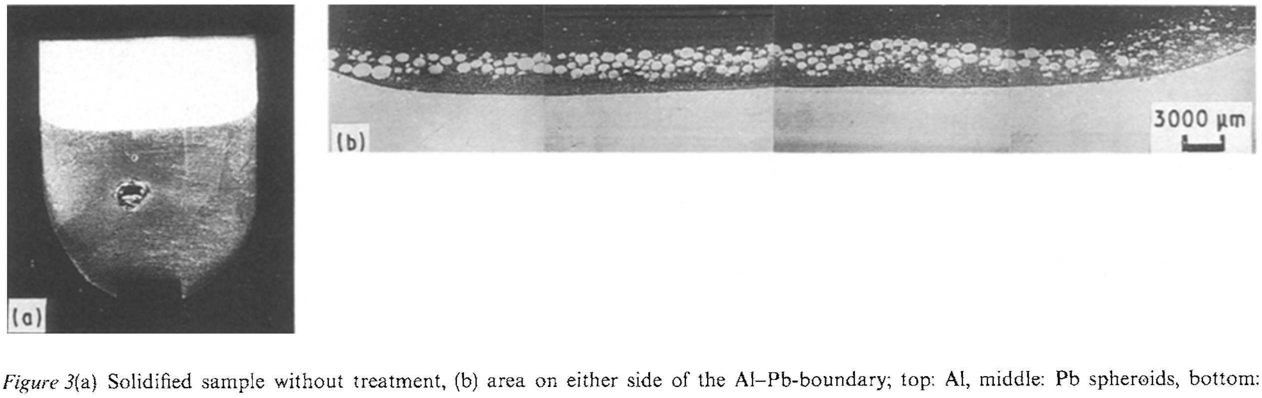

Figure 3(a) Solidified sample without treatment, (b) area on either side of the Al-Pb-boundary; top: Al, middle: Pb spheroids, bottom: sedimented Pb layer (SEM).

migration of the Pb spheroids is the thermo-solutal convection induced by the surface tension gradient which arises from either the temperature gradient or the concentration gradient or both [13, 14], the fluid motion as such being triggered off by the Lorentz forces superimposed on by the applied current.

During the cooling of the melt there is a temperature gradient between the crucible and the centre of the melt, and also between the liquid Pb and the hotter solidifying front on the Al side. In a separate experiment in which temperature measurements at a location just above the sedimented layer were undertaken, a continual fluctuation of the temperature was registered which goes to prove the assumption that this temperature gradient could have led to thermo-solutal convection. It is likely that the spheroids of lead which have nucleated at the crucible wall tend to migrate along the temperature gradient. Some of the lead droplets above a critical size, which have not been entrained by the aluminium matrix, shower downwards towards the sedimented lead surface. Fig. 4 shows the frequency of distribution of lead dispersoids entrapped in the matrix. The volume fraction of the dispersed phase on the Al side 25 mm away from the Al–Pb interface amounted to about 5%.

In the Pb-rich layer down below there are isolated aluminium spheroids of different sizes to be seen, preferentially adjacent to the Al–Pb boundary. In addition there are also fragmented dendrite segments, some of them enveloping lead particles in their centres (Figs 5 and 6). The spherical particle and the twisted raft-like figure were analysed in the energy-dispersive mode of X-ray analysis (EDXA) as Pb and Al, respectively. The fact that there are more Pb dispersions in the Al matrix than vice versa indicates that the gravitational sedimentation of Pb was possibly retarded by the commencement of crystallization of Al at 932 K. On the other hand the lighter aluminium spheroids and the fragmented trunks and arms of the dendrites float up to the area close to the Al–Pb boundary, depending on their velocity, before the onset of Pb solidification. Their unusual morphology can be attributed to the shearing action of the fluid body forces resulting in convection waves which cause the growing dendrite structure to collapse into bent and distorted rafts (Fig. 7).

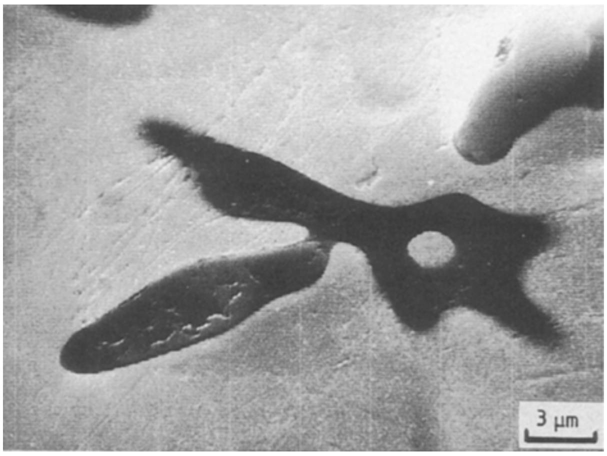

Figure 5 Broken dendrite raft and the enclosed Pb spheroid in Pb matrix (light), Al dendrite (dark) (SEM).

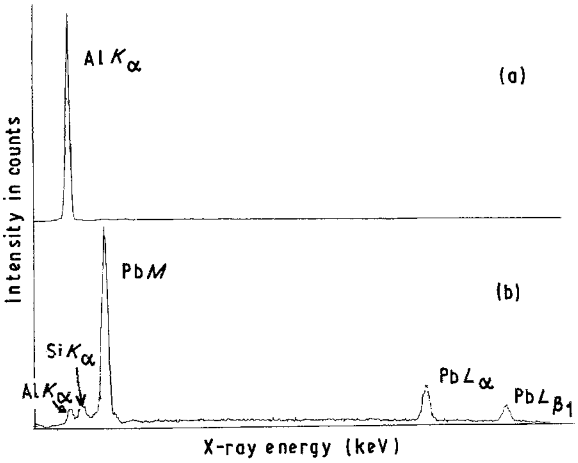

Figure 6 EDX-analysis of Fig. 5: (a) fragmented raft, (b) enclosed spheroid.

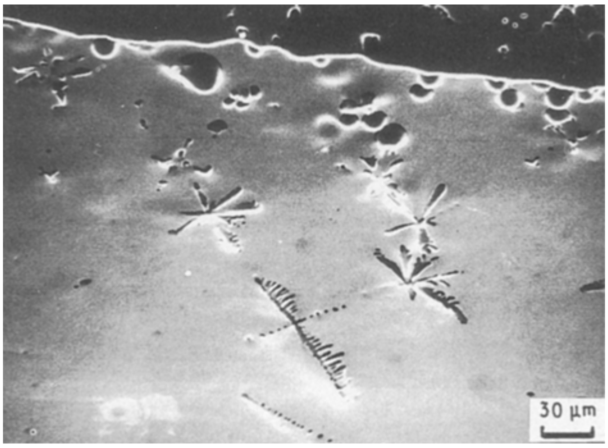

Figure 7 Broken dendrite rafts (SEM).

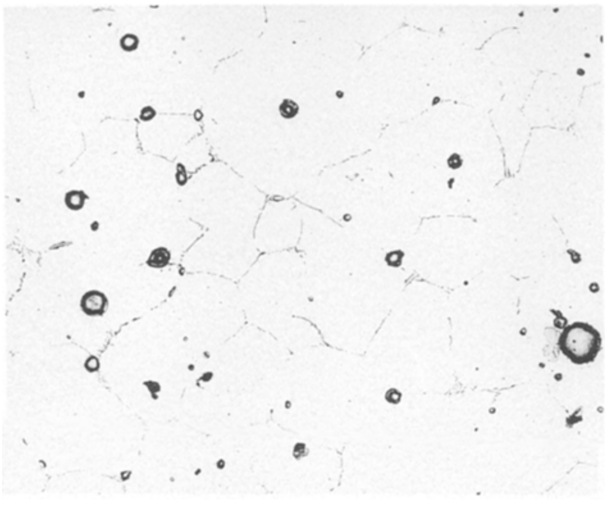

Figure 4 Pb dispersed in Al matrix.

Between the Al–Pb interface and the Pb spheroids, a layer of a network of fine-grain structure of aluminium could be observed. The grain-refining effect of the AlTiB master alloy is widely known [15, 16]. TiB_2

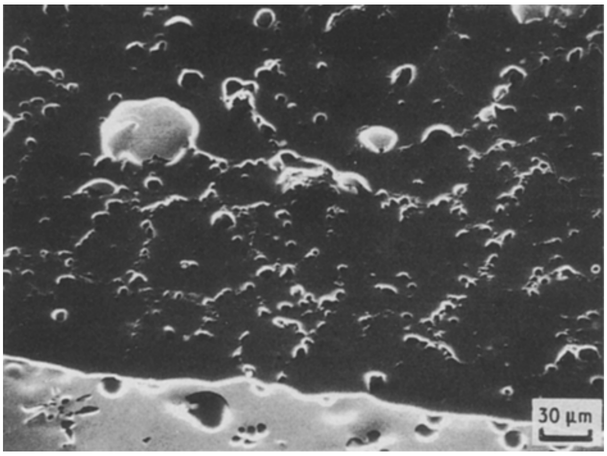

Figure 8 Region between the Pb spheroids and the Pb layer (SEM).

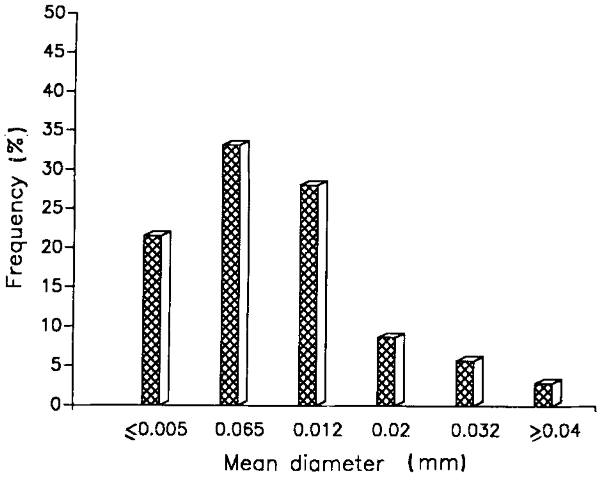

Figure 9 Size distribution frequency of Pb-rich dispersoids, 25 mm away from the sedimented Al–Pb interface.

clusters were identified in the grain centres as well as on the grain boundaries. The smaller liquid Pb droplets which find themselves close to the interface on the Al side, after having experienced a buoyancy effect resulting from the temperature gradient, are likewise rejected by the growing Al crystals towards the grain boundaries where they subsequently solidify. Fig. 8 shows Pb-rich dispersoids and titanium diboride clusters at the grain boundaries.

Some Al_3Ti plates and TiB_2 clusters were observed at the outer periphery of the sample. This confirms the fact that an azimuthal motion prevailed in the melt due to which some of the plates and the clusters were swept off to the outer edge of the melt just before the solidification commenced.

A size distribution analysis of the lead dispersions entrapped in the Al matrix was carried out about 25 mm from the Al–Pb boundary. Fig. 9 shows the distribution frequency of the dispersed phase in different size classes. Judging from the volume fraction of the dispersed phase, the alloy composition of the specimen at the measured site corresponded approximately to AlPb5.

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