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Rapid pressure solidification in an Al–Si alloy

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This study describes the effect of rapid pressure applied during solidification and the resulting microstructures and properties of an Al–1.7% Si alloy. The data on the microstructures and mechanical properties of the test alloy, together with the apparatus used to achieve rapid pressurization are discussed.

In this research, the pressure effects on microstructures of the test alloy were found to be significant. The mechanical properties such as yield strength and hardness, however, showed only minor differences in samples solidified under normal (1 atm) and pressurized conditions. This may be due to the competing effects of the dendritic and eutectic phases in the resulting structure. The results of this research suggest that high pressurization rate, applied during solidification actually achieves similar effects as high cooling rate in much larger dimensions of samples, thus, rapid pressure solidification may potentially offer an alternate method of obtaining the advantageous properties of rapidly solidified materials.

1. Introduction

Rapid solidification processing (RSP) has been a subject of active research since 1960 [1]. A great number of rapid solidification processing techniques have been developed, and superior properties have been found in materials produced by these methods. The research effort in this field has been largely restricted to the consideration of heat transfer and to the characterization of new microstructures and properties in relation to the cooling rate and degree of undercooling involved.

Among these RSP techniques, quenching is the most popular one [2]. In order to alter the crystalline phase, the cooling rate must be high enough to avoid the nucleation of stable phases. To achieve an adequate cooling rate, the heat extraction rate must be extraordinarily high. This condition limits the dimensions of products produced by the method of rapid quenching. Usually the material is formed in the shape of powder or foil with dimensions typically in the range of micrometres [3]. These dimensions are too small for most direct engineering applications. Although these products can be further consolidated by powder metallurgical methods, this consolidation leads to a general diminution of advantageous properties [4]. This severe limitation in size provides an incentive to search for an alternative method that would lead to materials with the same superior properties, yet of larger dimensions.

From a thermodynamic point of view, pressure and temperature are state functions. There are an infinite number of paths to obtain a final solid state from the liquid state [5]. In the proposed technique a rapid pressure increase is substituted for a rapid temperature decrease. The objectives of the present investigation are to demonstrate the feasibility of rapid pressure solidification (RPS) in producing materials with desired properties and to assess the advantages and limitations of this technique as compared to the conventional rapid cooling solidification (RCS) method. An Al–Si simple eutectic alloy was used as a test sample for this purpose. This choice was based on the simplicity of the alloy's phase diagram and its broad application in industry.

2. Experimental procedures

The maximum solubility of silicon in an Al–Si alloy is 1.6 wt % at atmospheric pressure (Fig. 1). Under high pressure this alloy is expected to have a much higher solubility of silicon. Theoretical calculations previously presented indicate that a maximum solubility of 2.5 wt % should be achievable at a pressure of

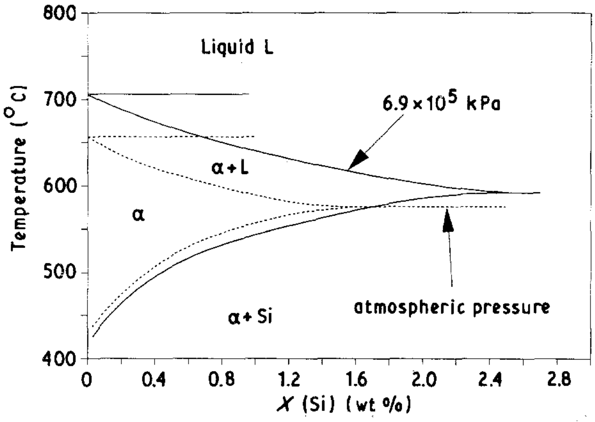

Figure 1 Phase diagram of Al–Si alloy at 6.9 × 10^5 kPa and atmospheric pressure.

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6.9×10^5 kPa [5]. An alloy with 1.7 wt% Si was chosen as a test composition to see whether high pressure would produce greater solid solubility and hence markedly different properties.

2.1. Materials

The alloy was prepared from an aluminium ingot (99.99%) and silicon lumps (99.9999%). The alloy was remelted and chemical analyses performed on samples taken from the top, middle and bottom to insure uniform composition. All samples tested (pressurized and control samples) were fabricated from this one starting stock.

2.2. Testing apparatus

A high speed hydraulic MTS testing machine was used to press the molten specimen in a pressure mould. The pressure mould was fabricated with a tapered hole bored into the centre. A tapered pressurization pin was also designed so that the tapered edges of the pin and mould could plastically deform and seal off the pressure during loading by the MTS machine (Fig. 2). A nominal pressure of 1.2×10^6 kPa was achieved in 30 msec. To monitor the temperature continuously, a k-type thermocouple was spark welded to the top of each mould. During calibration runs this temperature trace was compared with that of a thermocouple placed in the melt and the solidification temperature correlation determined, thus, the cooling rate is determined from this temperature–time plot.

To isolate the effect of a rapid pressure increase on solidification, it is desirable to have a control sample for each pressurized sample produced. Two specimens, therefore, pressurized and control, were made by the same process, the only differences being that the control sample was subjected to no load (pressurization) during solidification.

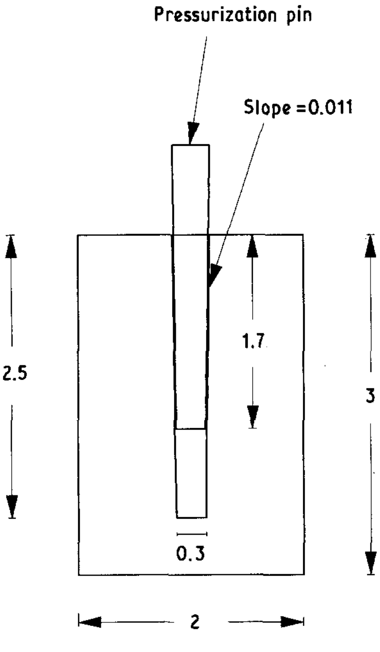

Figure 2 Schematic diagram of pressure mould (stainless steel) and pressurization pin (tool steel) (dimensions in inches, 1 inch = 2.5 cm).

The pressurized sample was mounted on the MTS machine immediately after removal from the furnace at a temperature of 750 °C. When the sample temperature dropped to 650 °C, it was pressurized at a desired rate. Then the sample was air cooled under load until the temperature reached 200 °C.

Microstructures of the pressurized and control specimens were examined by both optical and electron microscopy. Dendrite sizes of resulting samples were characterized relative to pressurization rate and cooling rate. Mechanical testing, including hardness tests and compression tests, was used to evaluate the improvement of properties.

3. Experimental results

3.1. Microstructures of pressurized and control samples

The alloy microstructures were found to be highly dependent on the solidification conditions. For a simple eutectic system, such as Al–Si, the eutectic structure and dendrite phase are dominant. Both of these structures can be related to pressurization and cooling rate.

3.1.1. Pressurization rate

A typical micrograph of the pressurized and control specimen is shown in Fig. 3. In general, the microstructure of the control specimen is primarily dendritic phase surrounded by a eutectic structure which may be complex regular or irregular flakes. For the pressurized specimens, different morphologies of the eutectic structures and intermetallic compounds were found. Three categories of differences between the pressurized and control group were identified.

(1) *Dendrite cell size.* The average dendrite cell size was measured using the statistical analysis. As shown in Fig. 4, the dendrite size of the pressurized specimen is smaller than that of the control over corresponding cooling rates. This agrees with the prediction that the pressurization rate should have the same effect as the cooling rate on dendrite size.

(2) *Eutectic structure.* The eutectic solidifies in a complex regular structure in the control specimens. This complex regular structure is only observed in the control samples. The pressurized specimens show only irregular flakes with a larger in-between spacing.

(3) *Intermetallic compounds.* Intermetallic compounds were found in all pressurized specimens but in only two (out of 12) of the control specimens. A much smaller volume fraction was found in these two control specimens than in the pressurized samples.

For the pressurized and control specimens the cooling rates were in the range of 50 to 69 °C min^{-1}. To ascertain that microstructural changes were due to pressurization rather than the small cooling rate differences, the following test was conducted. This test consisted of mounting identical specimens and solidifying the alloy in the MTS machine. One of the specimens was loaded to 50 kN and the other to a very small load of 0.1 kN in 10 msec (C1). Additionally for another sample the same load (50 kN in 10 msec) was

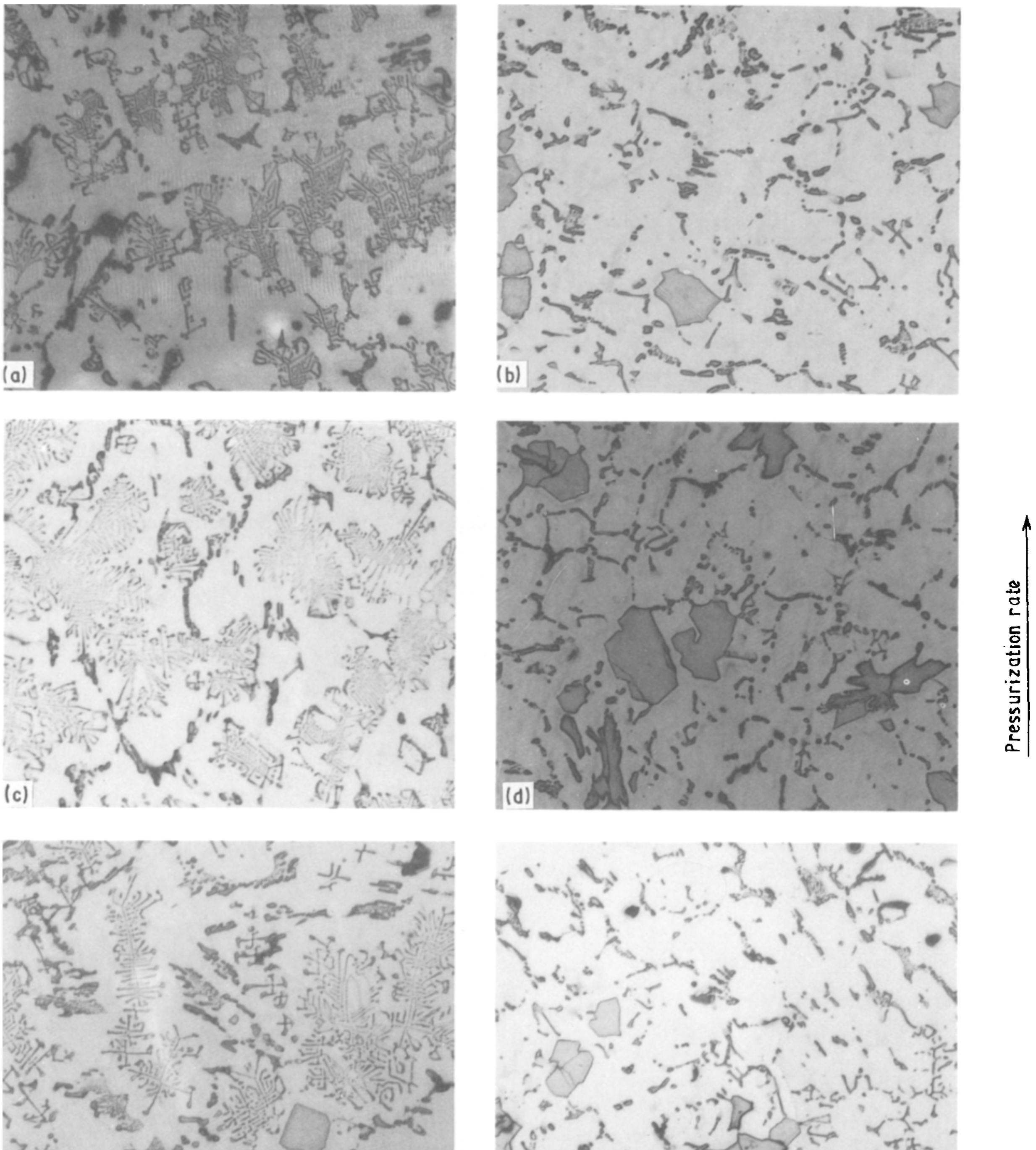

Figure 3 Micrographs of pressurized specimens ((b), (d), (f)) with corresponding control specimens, 200 × .

applied but two 30° channels were drilled into the mould to release the pressure, hence the sample beared no load (C2). The cooling rates were 50.8, 50.2, and 51.1 °C min^{-1}, respectively. These cooling rates are close enough to exclude the temperature effect on the resulting microstructures. As shown in Fig. 5, the morphologies of the two specimens under no pressurization are the same; however, they differ from that of the pressurized sample. The dendrite sizes are 21.24 and 21.52 μm, also different from the pressurized value of 14.96 μm, therefore, major differences in morphology are caused by pressure.

3.1.2. Cooling rate

Presumably, the control group should have the same morphology due to the hypothesis that the processing history is the same for all of the control specimens, however, slight differences in morphology were found due to sensitivity to the cooling conditions.

In order to isolate the temperature effect, three specimens which covered a much wider range of cooling rates than the normal pressurized and control specimens were prepared and examined. The cooling rates were 1.9, 52.8 and 99.8 °C min^{-1}, respectively.

Micrographs of these specimens are shown in Fig. 6.

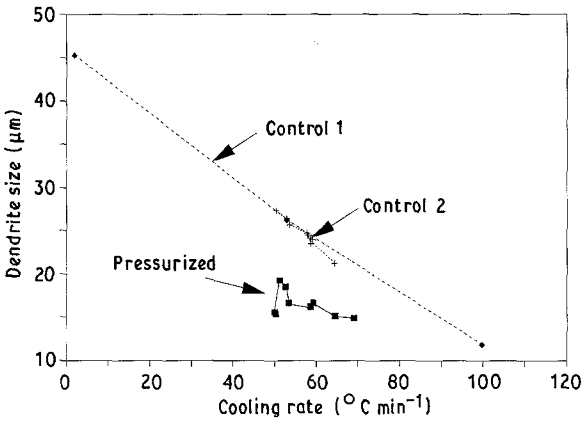

Figure 4 Plot of dendrite size against cooling rate (—— control 1, ● control 2, ○ pressurized).

Except for dendrite cell size and eutectic spacing the microstructures are quite similar. The dendrite cell sizes are 45.3, 26.2 and 11.8 μm. These results indicate a tendency of decreasing dendrite cell size with increasing cooling rate. These data (called control-1 in Fig. 4), together with all of the data from the normal control specimens (called control-2) are plotted in Fig. 4. The relationship between cooling rate and dendrite size of the pressurized specimens is also shown in Fig. 4. The dendrite size decreases as cooling rate increases; yet, dendrite sizes are typically 7 μm smaller than that of the control specimens in the same range of cooling rates. A somewhat smaller effect of cooling rate on the dendrite cell size in pressurized samples was noted.

3.2. Mechanical properties

3.2.1. Hardness

The microhardness of the dendrite phase, eutectic phase, and intermetallic compounds were measured separately. Among these three phases, intermetallic compounds are the hardest. The microhardness results are plotted in Fig. 7 with respect to the corresponding cooling rate for both pressurized and control specimens.

For the dendrite phase, the hardness is higher in the pressurized specimens than that of the control; for the eutectic phase, it is the reverse. The hardness of both phases is proportional to the pressurization rate (see Fig. 8), although the relationship is not very profound. The higher the pressurization rate, the higher the hardness that can be obtained.

3.2.2. Yield strength

The yield strength was measured from the compression tests. Although the data of the pressurized specimens show less dependence on dendrite size than that of the control, a Hall–Petch type relationship between yield strength and dendrite cell size exists as shown in Fig. 9.

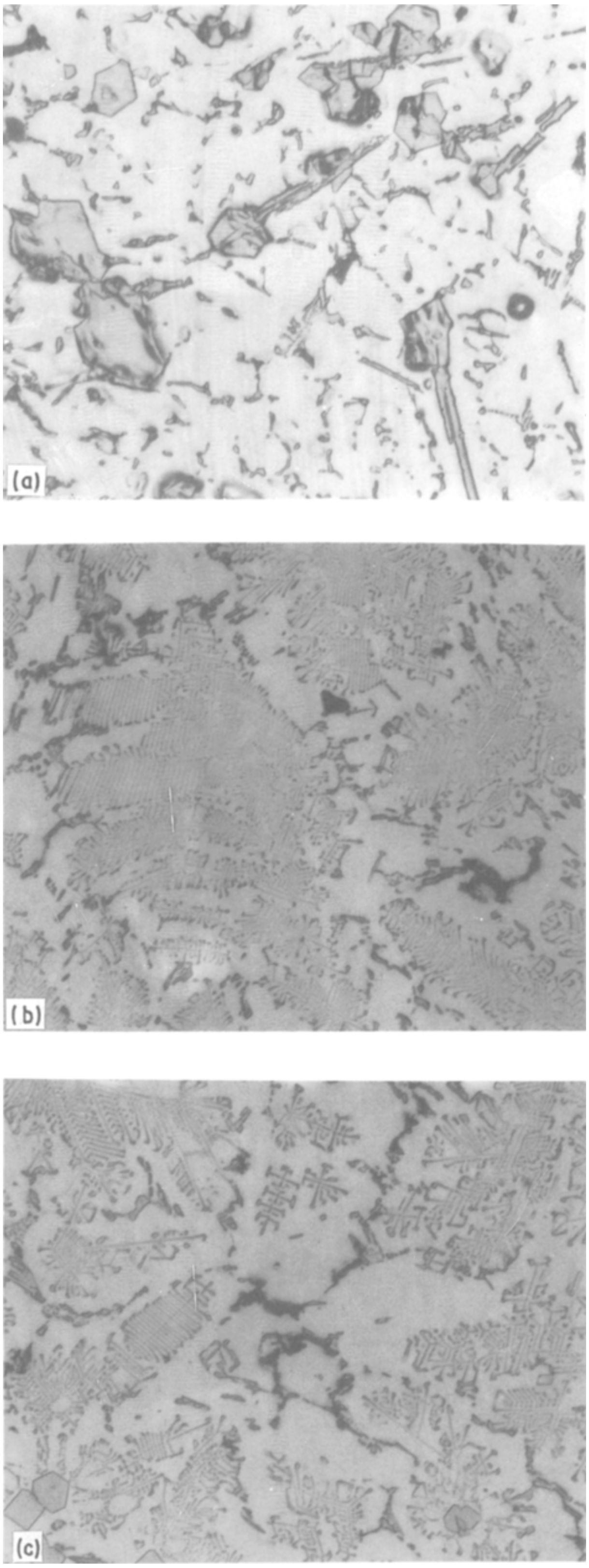

Figure 5 Micrographs of specimens showing pressure effect on morphology, 200 × (a) pressurized, (b) C1, (c) C2.

4. Discussion

4.1. Pressure effect

4.1.1. Microstructure

Generally dendrite cell size decreases when cooling rate increases. A somewhat similar trend was found for pressurization rate. The morphology of the pressurized sample is similar to that of the control sample,

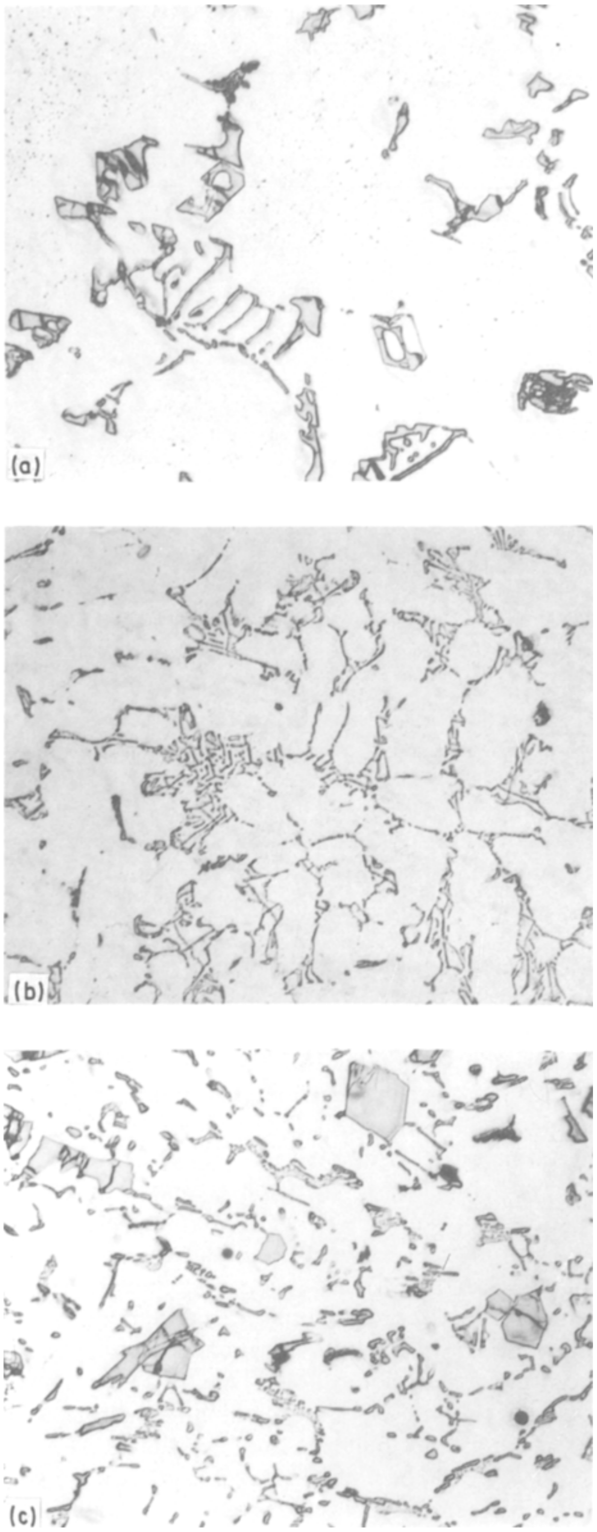

Figure 6 Micrographs of control specimens with cooling rates (a) 1.9, (b) 52.8, and (c) 99.8 °C min^{-1} respectively, 200 × .

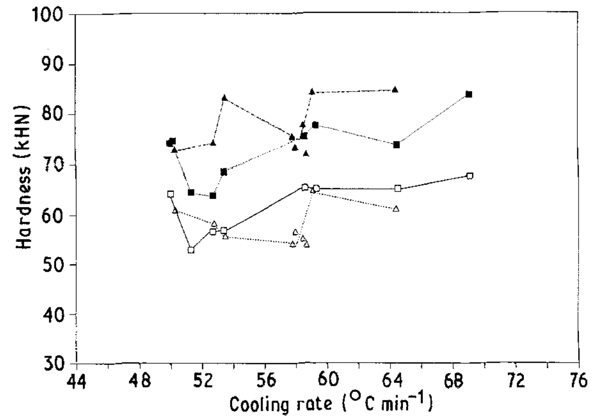

Figure 7 Plot of hardness against cooling rate (▲ control eutectic, ■ pressurized eutectic, □ pressurized dendrite, △ control dendrite).

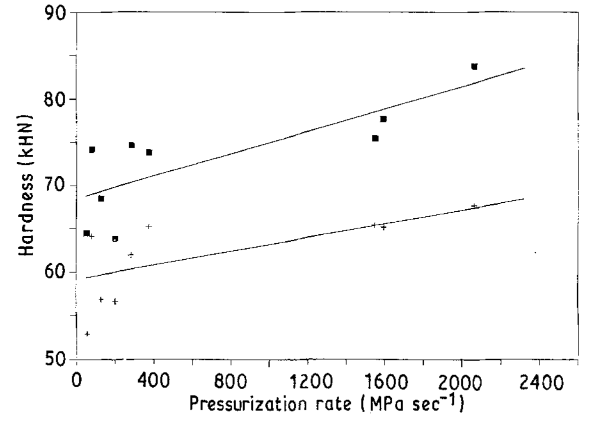

Figure 8 Plot of microhardness against pressurization rate in eutectic phase (■) and dendrite (+) phase.

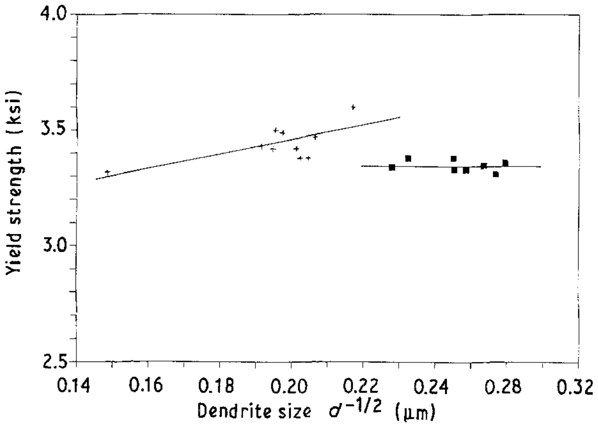

Figure 9 Plot of yield strength against dendrite size for both pressurized (■) and control (+) specimens.

however, the dendrite cell sizes of the pressurized specimens are smaller with a dispersed eutectic structure. It is confirmed that a high pressurization rate can decrease the dendrite size, as is also true for a high cooling rate. The dispersed eutectic structure is responsible for the change of yield strength and hardness. The yield strength of pressurized specimens is slightly smaller than that of control specimens. These results seem to be contrary to our prediction that higher solubility of silicon should achieve better mechanical properties. This may be explained by two reasons. First, the eutectic structure is dominant in determining the strength of an Al–Si alloy. For the pressurized specimens, the eutectic phase is more dispersed and the spacing between irregular flakes is much larger, therefore, the strength decreases due to this feature. Second, although the maximum solubility of silicon is 2.5 wt % under 6.9×10^5 kPa, the composition of the test alloy is 1.7 wt % of silicon. Thus the solubility only increases from 1.6 to 1.7% in the pressurized specimens. Any additional precipitation strengthening achieved by pressurization may not be significant.

The process of rapid pressure solidification uses rapid pressure increase instead of rapid cooling. The solidification time is in the range of a millisecond. During such a short time, diffusional process for eutectic growth is limited, therefore, the resulting structure is more dispersed with a larger spacing between irregular flakes. The nucleation of eutectic phase is also limited due to a small degree of undercooling; this also encourages a larger spacing between irregular flakes.

4.1.2. Hardness

The macrohardness of pressurized specimens is determined by the competing results of the eutectic and dendrite phases. For the dendrite phase, the hardness is higher in the pressurized specimens than in the control specimens. The opposite is true for the eutectic phase. The present data are not sufficient to decide the dominant criterion.

The hardness of the eutectic phase is greater than that of the dendrite phase in an Al–Si alloy. This is caused by silicon in the eutectic structure. The reason for the difference of hardness in the pressurized and control sample is explained as follows.

1) *Dendrite phase.* Higher solid solubility of silicon in the pressurized specimens (this is one of the principle advantages of RPS) is responsible for the microhardness increase.

(2) *Eutectic phase.* Finer (closer) eutectic spacing was found in the control specimens; this causes an increase in microhardness.

High pressure may thus result in higher solubility of silicon and improve the hardness of solid solution strengthened materials.

4.1.3. Yield strength

Previous investigators [6, 7] have shown that the Hall–Petch type relationship, $S = S_0 + kd^{-1/2}$, should describe the ultimate tensile strength and yield strength dependence on dendrite cell size in Al–Si eutectic alloys. The yield strength of pressurized and control specimens in this research also suggests such a relationship (see Fig. 9). The very flat slope of the Hall–Petch plot shows that the dependence of yield strength on dendrite size is not pronounced, however, two features may be observed. First, there is a higher dependence of yield strength on dendrite size of the control specimens than that of the pressurized specimens. Second, the yield strength of the control specimens is slightly greater than that of the pressurized specimens. This may be due to the differences in eutectic structure. For the control specimens, a complex regular structure is observed and eutectic spacing is smaller. The microhardness results show a higher hardness in the eutectic phase of the control specimens than that of the pressurized specimens. The finer spacing in the eutectic probably dominates strength determining factors, causing a higher yield strength in the control specimen.

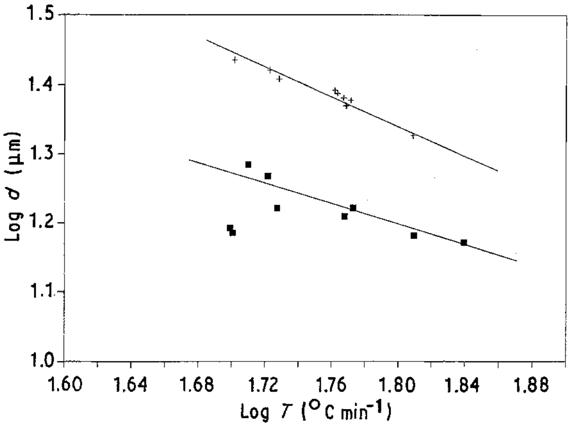

Figure 10 Dendrite size as a function of cooling rate for pressurized (■) and control (+) specimens.

4.2. Cooling rate effects

Dendrite secondary arm spacing and dendrite cell size have been related experimentally to cooling rate by power form as

$$d = A\dot{T}^{-c}$$

where typically $0.3 < c < 0.5$ [8]. Such a relationship exists as shown in Fig. 10. The constants of the pressurized and control specimens are 0.3 and 0.5, respectively, i.e. the pressurized specimens show less dependence on cooling rate.

4.3. Intermetallic compounds

It has been reported [9] that additional elements (such as iron, manganese, magnesium . . .) easily form intermetallic compounds in an Al–Si alloy. These compounds usually contribute to high strength and hardness.

An energy dispersive spectroscopy (EDS) analysis was performed to analyse the composition of each phase. The results show that aluminium, silicon, iron and chromium were detected in differing amounts for all phases in the selected specimens, although iron and chromium were present only in very small amounts. Impurities, iron as well as chromium may have come from the preparation process. Due to the existence of iron and chromium besides aluminium and silicon, the structure may be different from the simple Al–Si eutectic. It should be noted that pressurization increases the volume fraction of these impurity dominated precipitates which form.

5. Conclusions

The following conclusions were drawn from the experiments and analyses of this investigation.

(1) Rapid pressurization during solidification decreases dendrite cell size.

(2) Rapid pressurization during solidification alters the eutectic microstructure from complex regular to irregular flake.

(3) Dendrite cell size of both pressurized and control samples follow a $d = A\dot{T}^{-c}$ relationship.

(4) Macrohardness is determined by the competing results of the eutectic and dendritic phases of the

pressurized samples. For dendrite phase, the hardness is higher in the pressurized samples. The opposite is true for eutectic phase.

(5) A Hall–Petch type relationship, $S = S_0 + Kd^{-1/2}$, is observed for both pressurized and control samples.

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