JOURNAL OF MATERIALS SCIENCE 30 (1995) 452–458

The effect of small additions of scandium on the properties of aluminium alloys

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Additions of up to 1 wt% scandium have been made to Al, Al–Mg, Al–Mg–Ag and Al–Zn–Mg alloys and the effects on age-hardening and mechanical properties studied. Scandium levels up to 1% could be retained in solution at solidification rates of about 300 $K s^{-1}$. The precipitation of Al_3Sc at ageing temperatures in the range 563–593 K (290–320 °C) gave significant additional hardening. The low solubility of Sc in the solid state makes it difficult to obtain optimum hardening from Sc and other precipitating elements because of difficulties in solution treatment. The effect of deformation prior to ageing and the temperature-dependent mechanical properties are described.

1. Introduction

Scandium is sparingly soluble in aluminium in the solid state and supersaturated solid solutions decompose on ageing at elevated temperature, precipitating the Al_3Sc (L12) phase. This phase can form as a very fine dispersion of spherical particles, providing a considerable increase in strength to aluminium alloys.

The very high specific strengthening effect of small additions of scandium was first reported in the literature in 1971 in the form of a US patent [1]. Significant improvements in properties were found for Sc additions in the range 0.2 to 0.6 wt% (all compositions in this paper are given in weight percentage). The patent contains information on the properties of a wide range of alloys for a variety of mechanical and thermal treatments. In particular, the beneficial effect of deformation before ageing and the use of high ageing temperatures (e.g. 563 K (290 °C)) was noted. By way of example, an Al–5.25% Mg alloy containing 0.3% Sc had a yield strength of 365 MPa, more than double the strength of the Sc free alloy.

Russian work on Al alloys containing Sc appears in the literature from 1973 onwards [2–6] and was summarized in 1992 [7] where the optimum level of Sc and its interaction with other elements is discussed. The maximum useful amount of Sc that could be used was reported as 0.6% for alloys produced in the form of D.C. (Direct Chill) billet, rather higher than the maximum equilibrium solubility of 0.4%. In terms of the elements normally used for precipitation hardening in Al alloys it was noted that Sc did not form compounds with Mg, Zn, or Li but that its effect was very much reduced in alloys containing Cu and Si. The paper also notes the use of Sc in several Russian commercial alloys based on the Al–Mg, Al–Zn–Mg–Zr and Al–Li–Mg–Zr systems.

The most extensive recent work on alloys containing Sc has been on the binary alloys and on alloys containing Mg. Mg does not enter the precipitate structure and the strengthening effect of the precipitate is additive to the solution strengthening of Mg and any dislocation substructure that may be present. This work is discussed in more detail below.

1.1. Al–Sc binary alloys

Fig. 1 summarizes some of the data on the yield strength of binary alloys taken from the literature; some results from the present work are included for comparison and discussed, in more detail, later. Maximum strength and hardness is found in these alloys for ageing temperatures in the range 523 to 623 K (250

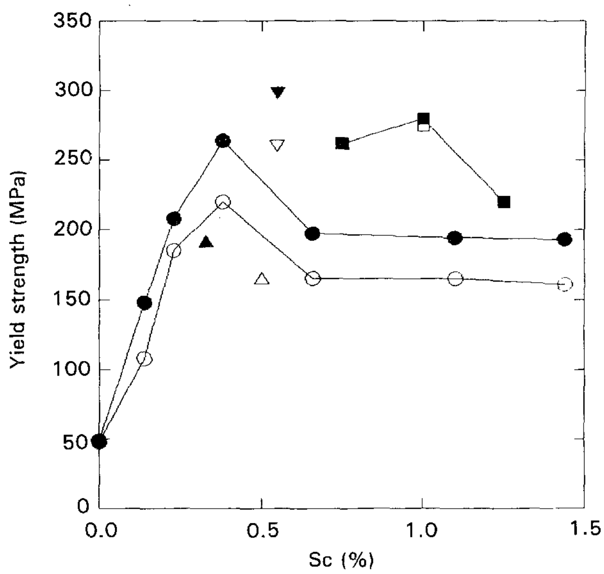

Figure 1 Yield-strength data for binary aluminium–scandium alloys from the literature and the present work. Sourçes were as follows. Willey [1]: (○) as-cast and aged at 563 K (290 °C); (●) cold rolled 80% before ageing. Sawtell and Jensen [9]: (▽) as-cast and aged at 563 K (290 °C); (▼) warm-rolled, cold-rolled and aged at 563 K (290 °C). This work: (□) as-cast, aged at 593 K (320 °C); (■) cold-rolled 83%, aged at 593 K (320 °C). Drits *et al.* [4]: (△) homogenized at 773 K (500 °C), aged at 523 K (250 °C). Torma *et al.* [8]: (▲) solution-treated at 913 K (640 °C), aged at 583 K (310 °C).

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to 350 °C). The results of Sawtell and Jensen [9] and the present work show the additional strengthening that may be obtained with higher supersaturations of Sc. The alloys which received a high-temperature homogenization or solution treatment did not show as great a strengthening effect (compare the results of Drits *et al.* [4] and Torma *et al.* [8] with those of Willey [1]). The results of Willey and those of Sawtell and Jensen [9] show the benefit of cold-working the cast alloys prior to ageing, although only a small effect was seen in the present work, where the supersaturation of Sc is higher, an observation we shall return to later.

1.2. Al–Mg–Sc alloys

Sawtell and Jensen [9] have made an extensive recent study of alloys containing Mg and an addition of 0.56% Sc. The best combination of results was reported for alloys containing 4% Mg after various initial treatments. In the as-cast and aged condition the yield strength was 310 MPa and this rose to 380 MPa with a warm-rolling treatment prior to ageing, and to 414 MPa with a cold-rolling stage following the warm rolling. An alloy containing 6% Mg and 0.54 Sc, following a similar treatment, had a strength of 433 MPa. Alloys with more than 4% Mg, however, were found to be brittle and were not extensively studied. The strength of the alloys fell quite sharply with test temperature, despite the high ageing temperature, and indeed the deformed and aged alloys were found to be superplastic. This finding led to a further patent [10].

1.3. The present work

The work reported here had several objectives. Firstly, it was intended to see if more Sc could usefully be taken into solution by solidifying at a higher rate and, secondly, to examine the effects of other alloying elements. These included Ag, following the well-known work of Polmear and Sargeant [11] on the role of Ag in promoting age-hardening in alloys containing Mg, and Zn to examine the additive effect of Sc on the high-strength 7000 series alloys. The effect of deformation prior to ageing has also been studied to examine its effect on elevated-temperature strength and superplasticity.

2. Experimental procedure

Several Al–Sc, Al–Sc–Mg, Al–Sc–Mg–Mg and Al–Sc–Mg–Zn alloys were made using 99.9% Al, an Al–5% Sc master alloy and either pure metal or master alloy additions of the other elements. The alloys were chill-cast in a water-cooled Cu mould to a thickness of 4 mm which gave a solidification rate of 300 $K s^{-1}$. At this solidification rate it was possible to retain close to 1% Sc in solution, since very little second phase could be detected by optical or electron microscopy.

The ageing of some samples was studied in the as-cast state and others were cold-rolled prior to ageing, mostly to a reduction of 83%. The effect of different reductions was also studied. Ageing response was initially recorded by Vickers hardness (H_v) testing and appropriate samples were taken for tensile or compression testing. Microstructures were studied by optical and electron microscopy.

3. Results and discussion

3.1. Al–Sc binary alloys

Initial ageing studies were made on samples of the cast alloys cold-rolled 83% prior to ageing at 563 K (290 °C). Fig. 2 shows the age-hardening response for alloys containing from 0.5 to 1.25% Sc. Peak hardness is reached after about 800 min ageing at this temperature. The highest hardness (125) was found for the alloy containing 1% Sc. Significant second phase was found in the 1.25% Sc alloy, reducing its hardening capacity. The results show that, with a solidification rate of 300 $K s^{-1}$, approximately 1% Sc can be retained in solid solution and give additional hardening when precipitated by an appropriate ageing treatment.

At a later stage in the work, the hardening of the as-cast alloy was measured for an ageing temperature of 593 K (320 °C) and the results are shown in Fig. 3. The maximum hardness, after 100 min ageing, is 112 for the 1% Sc alloy. Given the higher ageing temperature for these samples, it would appear that cold work prior to ageing has only a small effect for high Sc contents, an observation we discuss further below.

3.2. Al–Sc–Mg alloys

Following the work of Sawtell and Jensen [9], an Mg content of 4% was chosen since higher levels showed evidence of brittleness in addition to higher strength. The ageing response of alloys with different Sc contents aged at 563 K (290 °C) after 83% deformation is

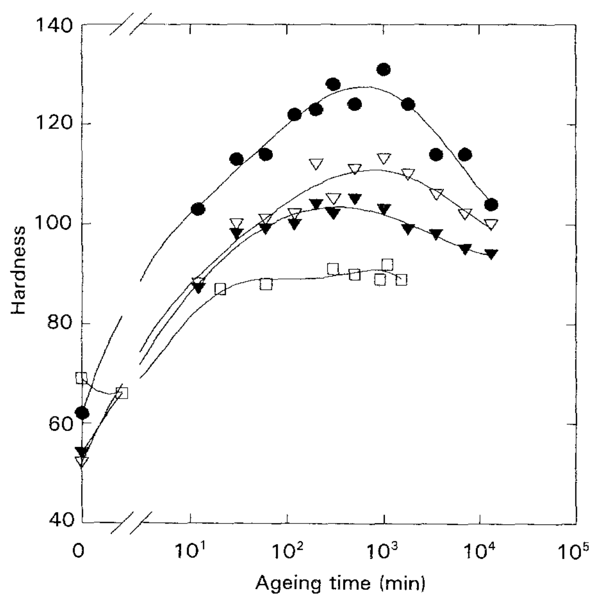

Figure 2 Age-hardening response of binary Al–Sc alloys aged at 563 K (290 °C) after 83% deformation by cold-rolling. Sc (wt %): (▼) 0.5, (▽) 0.75, (●) 1.00, (□) 1.25.

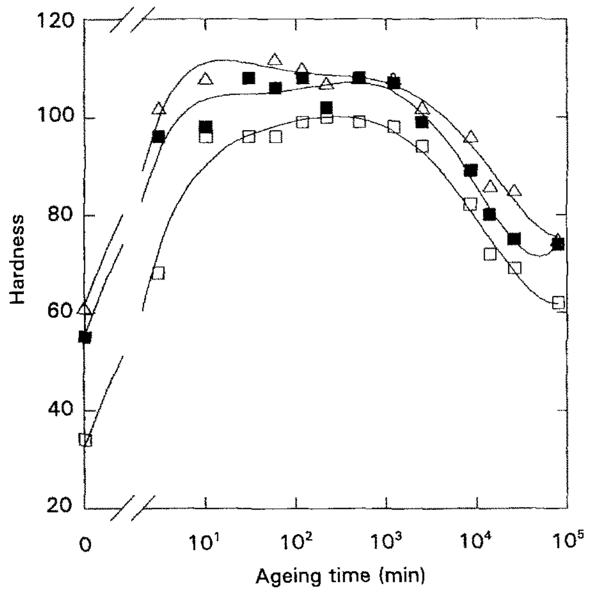

Figure 3 Age-hardening of as-cast binary Al–Sc alloys at 593 K (320 °C). Sc (wt %): (△) 0.5, (■) 1.00, (□) 1.25.

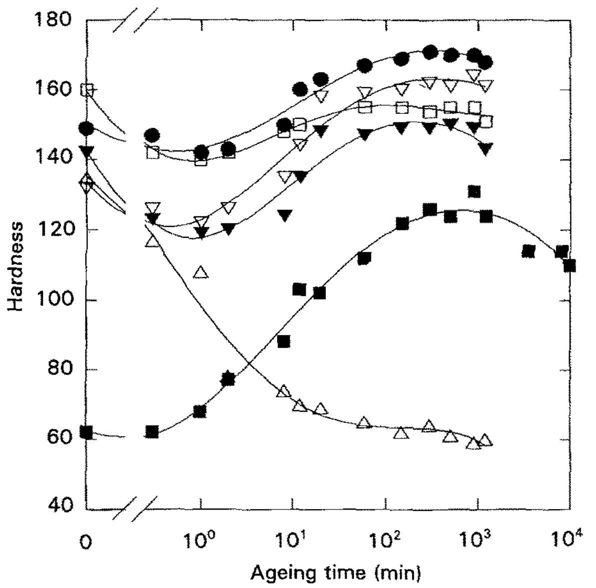

Figure 4 Age-hardening response of Al–4% Mg alloys containing Sc after ageing at 563 K (290 °C) after 83% deformation by cold-rolling. (■) 1%, Sc–(△) 4% Mg, (▼) 4% Mg–0.5% Sc, (▽) 4% Mg–0.75% Sc, (●) 4% Mg–1.00% Sc, (□) 4% Mg–1.25% Sc.

shown in Fig. 4 together with the results for the binary Al–1% Sc and Al–4% Mg alloys. A Sc addition of 1% to the Al–4% Mg alloy again shows the greatest hardening response with a maximum of 171 after 300 min ageing.

3.3. Al–Sc–Mg–Ag alloys

Silver promotes age-hardening in the Al–Mg system [11] and so alloys were made to see whether the effect could be additive to the hardening from scandium. Fig. 5 shows the effect of adding 0.4% Ag to alloys containing 1% Sc and 4, 5 and 7% Mg. The as-cast alloys were again deformed 83% by rolling prior to ageing at 320 °C. It should be noted that this ageing temperature is very high for the Al–Mg–Ag system, where hardening is seen for temperatures below about

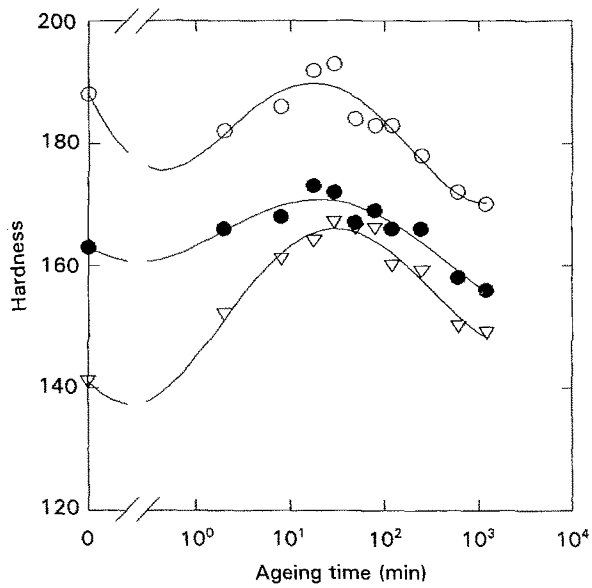

Figure 5 Age-hardening of Al–Mg–Ag–Sc alloys at 593 K (320 °C) following 83% cold-rolling: (▽) 4% Mg–1% Sc–0.4% Ag, (●) 5% Mg–1% Sc–0.4% Ag, (○) 7% Mg–1% Sc–0.4% Ag.

473 K (200 °C) [11]. The hardening of the 4% Mg alloy is similar to that of the equivalent silver-free alloy ($H_V = 165$ compared with 168 for the Ag-free alloy) and so it might be concluded that Ag is having no effect.

It is interesting to note the additional hardening with higher Mg contents, with a peak hardness of 192 for the Al–7% Mg–1% Sc–0.4% Mg alloy. A silver-free alloy of this Mg content was not made since Sawtell and Jensen [9] had noted very low ductilities in alloys containing over 4% Mg. Further work may be fruitful in this area.

Attempts with different heat treatments to obtain a hardening effect from the Ag addition were not successful. Solution treatment at 753 K (480 °C) prior to ageing produced a hardening response, similar to that of Al–Mg–Sc alloys, but the effect of Sc was largely lost. A low-temperature ageing treatment after the initial treatment to precipitate Sc also produced no additional hardening. We have yet to find heat treatments which combine the effects of Sc and Ag.

3.4. Al–Zn–Mg alloys

Willey [1] reported additional strengthening from Sc in this system and Elagin *et al.* [7] noted that Sc does not react with Zn or Mg and so an additive effect is possible. An alloy containing 6% Zn and 2% Mg was made with and without an addition of 1% Sc. Age-hardening was studied at 408 K (135 °C) after a number of initial treatments. There was no deformation prior to ageing.

The Sc-free alloy reached a peak hardness of 154 after ageing at 408 K (135 °C) following a standard solution treatment of 1 h at 748 K (475 °C) and water quenching. The alloy containing 1% Sc had a maximum hardness of 152 after a similar treatment, implying no additional hardening by Sc. Lowering the solution treatment temperature to 673 K (400 °C) led to a peak hardness of 164, indicating some worthwhile additional strengthening. This hardness would be approximately equivalent to a tensile strength of 500 MPa, which compares with a value of 440 MPa for the Sc-free alloy.

It is difficult to define a set of heat treatments which suits both the precipitation of Al_3Sc and the lower-temperature precipitating phases. Solution treatment is required to achieve effective hardening by the phases which form at lower temperature; however, solution treatment also causes precipitation of Sc as coarse, and non-hardening, particles of Al_3Sc.

3.5. Mechanical properties of Al–Sc and Al–Sc–Mg alloys

Mechanical properties were measured in tension on non-standard strip samples which were rolled prior to ageing. As-cast alloys were tested by compressing small cylinders. Fig. 6 shows the variation in yield strength and tensile strength for binary and ternary alloys aged to peak hardness at 593 K (320 °C) after cold-rolling 83%. The results of Sawtell and Jensen [9] for samples aged at 563 K (290 °C) are also shown. The binary alloy containing 1% Sc has a yield strength of 290 MPa and a tensile strength of 325 MPa. The addition of 4% Mg raises the yield strength to 410 MPa and the tensile strength to 530 MPa. In the present work there is a greater difference between yield and tensile strength than that reported by Sawtell and Jensen, and this may be accounted for by differences in mechanical and thermal treatment; otherwise the two sets of results are comparable, with the advantage of increasing the Sc level to 1% apparent from the present work.

Total elongation values are shown in Fig. 7. It appears that there is a small increase in elongation with Sc content for the binary alloys whilst there is a decrease for the ternary alloys. The ductility of the ternary alloys is reasonable for alloys with a high yield strength.

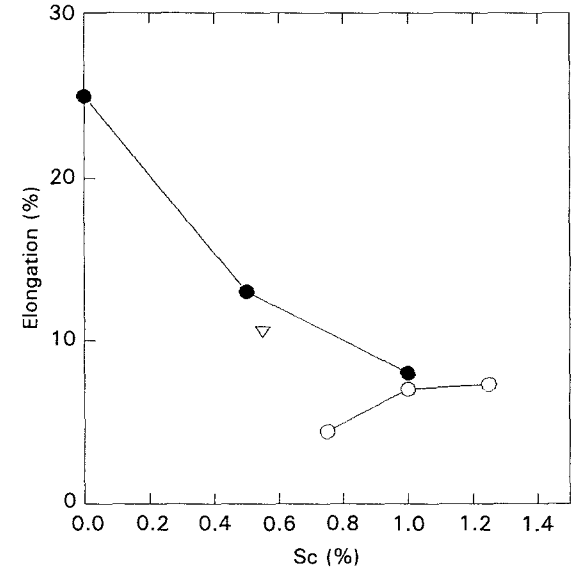

Figure 7 Total elongation results for Al–Sc and Al–Mg–Sc alloys aged to peak hardness at 593 K (320 °C) following 83% cold deformation: (○) 1% Sc, (●) 4% Mg–1% Sc, (▽) a similar alloy from Sawtell and Jensen [9].

3.6. Temperature dependence of mechanical properties

The high ageing temperatures used for the precipitation of Sc suggested that the elevated-temperature mechanical properties might be attractive, as the precipitate structure should be stable at temperatures in the vicinity of 473 K (200 °C). Fig. 8 shows the variation of tensile yield strength with temperature for alloys rolled prior to ageing, as well as data for the compressive yield strength of as-cast alloys of similar

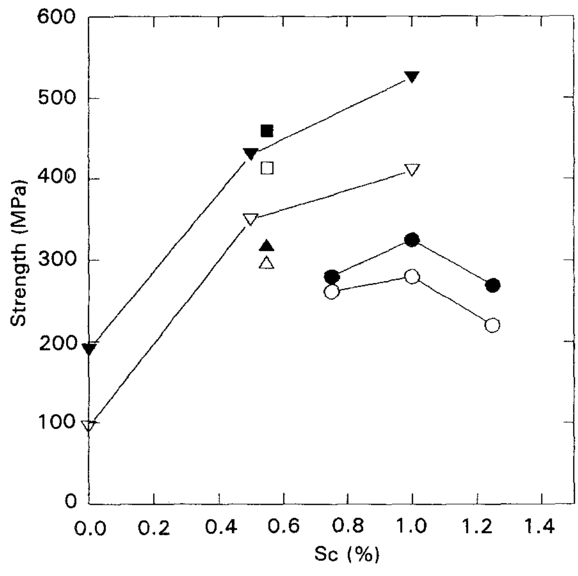

Figure 6 Yield (open symbols) and tensile (closed symbols) strength of Al–Sc and Al–Mg–Sc alloys aged to peak hardness at 593 K (320 °C) following 83% cold-rolling: (○, ●) 1% Sc, this work and (△, ▲) from Sawtell and Jensen [9]; (▽, ▼) 4% Mg–1% Sc, this work and (□, ■) from Sawtell and Jensen [9].

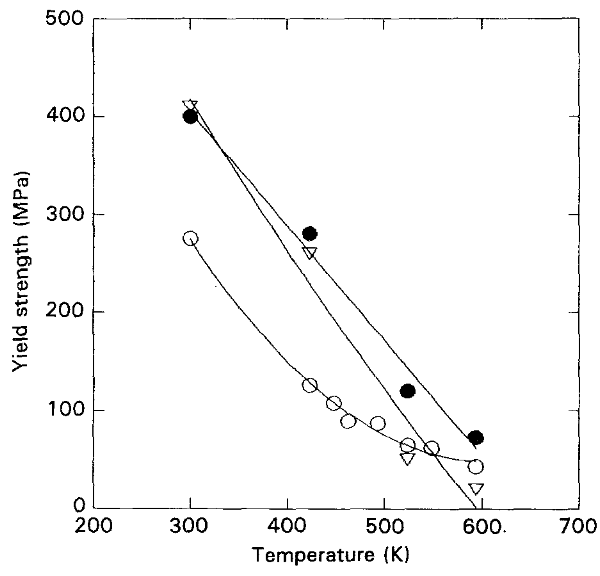

Figure 8 Temperature dependence of the yield strength for Al–Sc and Al–Mg–Sc alloys aged to peak hardness at 593 K (230 °C), measured in tension for cold-rolled samples and in compression for as-cast samples: (○) 1% Sc as cast, (●) 4% Mg–1% Sc as cast, (▽) the same alloy 83% cold-rolled.

composition and ageing treatment. The yield strength of the deformed and aged alloys falls quite sharply with temperature, indeed the decline is greater than for other age-hardening alloys intended for high-temperature use such as 2219-T851. As reported by Sawtell and Jensen [9], and also in an earlier paper by the present authors [12], the deformed and aged alloys become superplastic at quite low temperatures as a result of the fine sub-grain size stabilized by the fine Al_3Sc dispersion.

This observation led to a study of the as-cast alloys. It can be seen that the room-temperature strength is little affected by the deformation step (in contrast to the observations of others studying alloys of lower Sc content) but that the undeformed alloys are considerably stronger at higher testing temperatures, reflecting an absence of superplastic behaviour. These results show that deformation is not required to achieve effective age-hardening at high supersaturations of Sc. We must comment, however, that the need to cast in thin sections and the inability to restore properties through solution treatment might severely limit the application of alloys, except perhaps via superplastic forming of strip-cast and cold-rolled sheet. Although superplasticity did not occur in the as-cast alloys the elevated-temperature properties were still not as good as those of some other age-hardening alloys.

3.7. Optimum levels of plastic deformation prior to ageing

The observation of satisfactory strengthening at ambient temperature in the as-cast state led to a study of the effect of deformation on ageing behaviour. In Fig. 9 it can be seen that the age-hardening response is strongly dependent on the rolling strain; however, the hardening in the as-cast alloy is superior to that in the sample that received 15% rolling deformation. We will return to this observation when we discuss the microstructures.

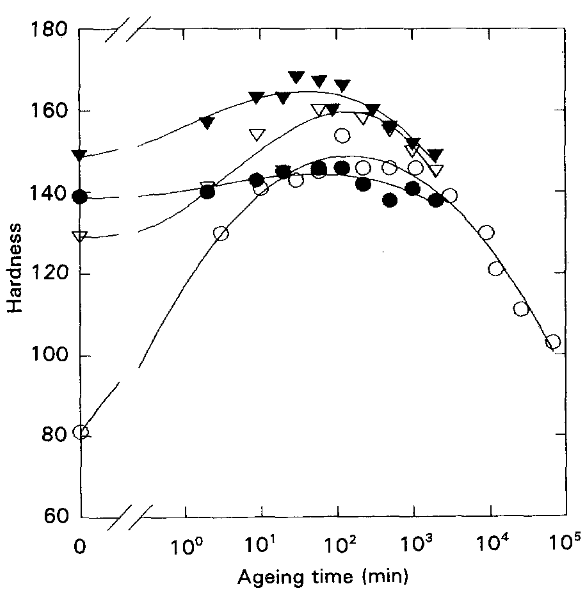

Figure 9 Effect of rolling deformation before ageing on the hardening response of an Al–4% Mg–1% Sc alloy aged at 593 K (320 °C): (○) as cast, (●) 15% deformation, (▽) 40%, (▼) 83%.

3.8. Microstructures

The microstructure of the 4% Mg–1% Sc alloy after cold-rolling to 83% and ageing to peak hardness (60 min) at 593 K (320 °C) is shown in Fig. 10. The fine 1 μm subgrain structure is evident but the precipitate structure could not be resolved using conventional TEM techniques. After over-ageing for 5500 min at 593 K (320 °C) coherent precipitates about 15 nm in diameter are visible and the subgrain structure has coarsened (Fig. 11). The strain-field contrast was analysed using the method described by Ashby and Brown [13, 14] and this gave a value for the elastic mismatch at the interface of 0.017 ± 0.02; this is a very high value for a coherent interface, but close to that expected from the lattice parameter difference between the matrix and Al_3Sc.

The sample deformed 15% by rolling showed a somewhat different structure (Fig. 12) showing evidence of discontinuous precipitation of the Al_3Sc. This precipitation behaviour is responsible for the poor ageing response of the lightly deformed samples. A

Figure 10 Microstructure of the Al–4% Mg–1% Sc alloy cold-rolled 83% prior to ageing to peak hardness at 593 K (320 °C).

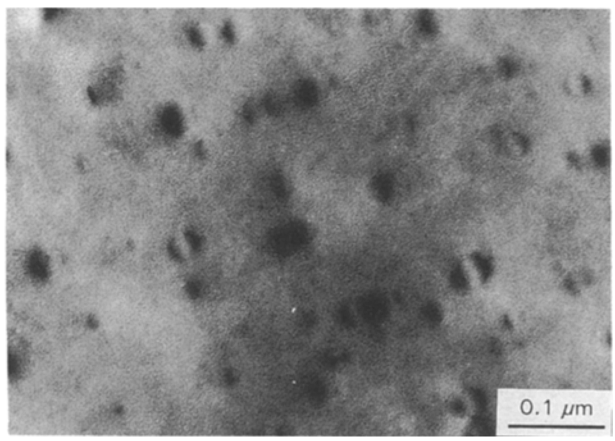

Figure 11 Microstructure of Al–4% Mg–1% Sc alloy after over-ageing for 5500 min at 593 K (320 °C). Coherent precipitates about 15 nm in diameter.

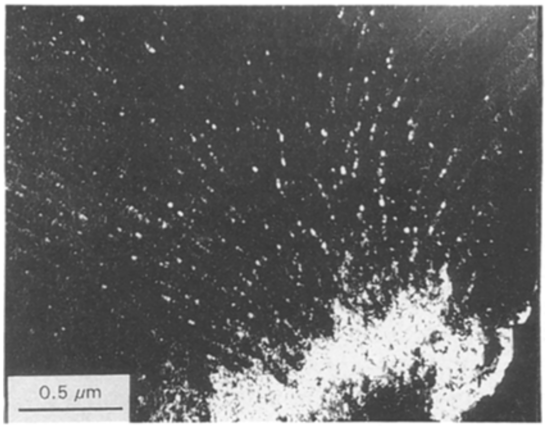

Figure 12 Microstructure of a similar sample to that of Fig. 11 but which had a deformation of only 15% prior to ageing. Discontinuous precipitation. (Dark field image.)

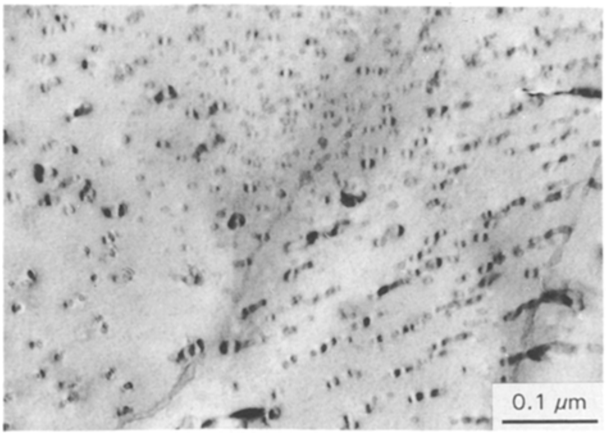

Figure 13 Another region of the sample in Fig. 11. Note some alignment of precipitates suggesting discontinuous precipitation, but note also coherency contrast at all precipitates.

careful study of more highly deformed samples also showed a tendency for precipitate alignment (Fig. 13), implying precipitation on temporarily arrested mobile boundaries. An important point, however, is that whatever the nucleation mechanism the precipitates are coherent with the matrix after the boundaries have passed, and the precipitate is more uniform throughout the matrix, the higher the deformation before ageing.

3.9. Precipitation of Al_3Sc

We have not made a detailed study of precipitation mechanisms in these alloys but a few points are made here which appear consistent with the observations. The effectiveness of prior deformation in facilitating the ageing response in previous work is associated with the nucleation of Al_3Sc. The mismatch between Al_3Sc and the matrix is large for coherent nucleation and it is clear that, near dislocations or subgrain boundaries, strain fields assist nucleation by reducing the strain energy required to establish a nucleus. None the less, once nucleated the precipitates appear to grow coherently with the matrix and there is no evidence for interfacial dislocations at long ageing times. In the present work we have found that as-cast alloys have a strong ageing response and conclude that coherent nucleation is just possible under the very high supersaturations present in alloys with 1% Sc. Light deformation seems to provide the conditions (mobile subgrain boundaries) for the kinetics of discontinuous precipitation to be more rapid than for homogeneous nucleation. When the dislocation density is very high, precipitation occurs uniformly throughout the structure because of the uniform high density of dislocations, a characteristic of Al–Mg alloys; the subgrain structure develops only slowly and the boundaries are pinned by the precipitates.

3.10. Strengthening mechanisms

The increase in strength due to the presence of Al_3Sc precipitates is about 240 MPa in the case of binary Al–1%Sc alloy and about 310 MPa for the Al–4% Mg–1%Sc alloy. These increases may be converted to approximate shear strength increments by dividing by the average Schmidt factor, 3.1. An explanation is, therefore, required for increases in the shear strength of from 80 to 100 MPa when 1%Sc is added.

Strengthening mechanisms in precipitation-hardened alloys were critically reviewed by Ardell [15] in 1985. There are a number of possible contributions which may be divided into those which dominate when dislocations cut through the particles and those which are effective when dislocations avoid the particles. In general, small coherent precipitates are cut by moving dislocations; the strength rises with particle size until, at some critical particle size, the mechanism changes to particle avoidance, and the strength becomes inversely proportional to the size.

Torma *et al.* [8] observed dislocation loops in Al–Sc alloys aged to peak hardness and deformed, leading to the suggestion that the avoidance mechanism was operating. If this were the case then the measured strengths are readily accounted for because of the fine dispersion. It is however unlikely that such small particles are strong enough to withstand being cut by the dislocations. Further, dislocation pairs were occasionally observed in under-aged samples in the present work, indicative of the cutting of small, ordered particles.

Significant contributions to the shear strength of the alloy when the precipitate particles are cut will come from the elastic coherency strains at the particle–matrix interface and from the work done in creating antiphase boundaries in the ordered particles. To make approximate calculations of the increase in strength ($\Delta\tau$), it is necessary to make some assumptions and approximations:

(i) The shear moduli G of Al_3Sc and the matrix are similar and are about 23 000 MPa.

(ii) The slip system in the matrix is $\{1\,1\,1\}\langle 1\,1\,0\rangle$ with a Burgers vector $\mathbf{b} = a\sqrt{3/2}$.

(iii) The lattice parameter a of the matrix is 0.404 nm and that of Al_3Sc is 0.410 nm, giving a $\Delta a/a$ of 0.015.

(iv) The particle size at peak hardness was not readily measured; the particle radius r was 7.5 nm when very overaged. An approximation using coarsening theory leads to a value of about 3 nm at peak hardness. The analysis which follows is not very sensitive to the precise value.

(v) Al_3Sc is fully precipitated with a volume fraction (f) of 0.023.

3.10.1. Contribution from the antiphase boundary energy

An equation which takes account of particle cutting by paired dislocations is

$$\Delta\tau = 0.81\gamma/2\mathbf{b}[(3\pi f/8)^{1/2} - f]$$

To calculate the increase in strength a value for the antiphase boundary energy γ is required. We are not aware of a measurement of the energy for Al_3Sc but we can make use of an approximation by Marcinkowski *et al.* [16] where

$$\gamma = 1.41\, k\, T_c/a^2$$

T_c is the order–disorder transformation temperature of Al_3Sc, assumed to be the melting point (1390 K) and k is the Boltzmann constant. This gives a value of 0.18 J m^{-2}, a little lower than the value calculated for the crystallographically similar γ'' phase in nickel alloys.

Using this value, the increase in shear strength due to the antiphase boundary energy of the particles is 30 MPa at peak hardness.

3.10.2. Coherency strengthening

Ardell [15] gives an equation for coherency strengthening in underaged alloys:

$$\Delta\tau = Const.\, (\varepsilon G)^{3/2}(rf\mathbf{b}/T)^{1/2}$$

and notes that similar equations were derived by many earlier workers, differing only in the value of the constant which lies between 2 and 3. We shall adopt the value used by Ardell, 2.6. The mismatch, ε, may be approximated by 2/3 $\Delta a/a$. The line tension, T, of the dislocation is often taken as 0.5 $G\mathbf{b}^2$ as an average for the values of pure edge and pure screw dislocations. It is appropriate, in the present analysis, to use the limiting value for the dislocation which finds the cutting process most difficult. This is the screw dislocation, and in aluminium the line tension for a pure screw dislocation is 0.14 $G\mathbf{b}^2$. Substituting values into the equation gives an increase in strength due to coherency strains of 48 MPa in the peak aged condition.

Ardell also gives an expression for the upper limit if dislocation flexibility is ignored:

$$\Delta\tau = 1.84\, G\varepsilon f^{1/2}$$

which leads to a value of $\Delta\tau$ of 64.3 MPa.

Combining the contributions from coherency and order hardening gives an overall increase in the shear strength due to the Al_3Sc particles of from 78 to 94 MPa, similar to the increase measured experimentally.

4. Conclusions

1. Scandium levels up to 1% can be retained in solution in aluminium alloys using solidification rates of 300 K s^{-1} or higher.
2. Additions of 1% scandium provide a greater strengthening effect than lower levels.
3. The high supersaturation promotes a more uniform precipitate distribution, reducing the need for heavy cold-deformation before ageing.
4. Scandium is very effective as an addition to Al–Mg solid-solution alloys. Additions to alloys containing other precipitating elements is less effective because of difficulties in finding compatible solution treatment and ageing temperatures. Of alloy systems for further consideration, the Al–Zn–Mg system shows promise.
5. High ageing temperatures in excess of 573 K (300 °C) do not lead to alloys with superior elevated-temperature properties. In Al–Mg–Sc alloys heavily deformed prior to ageing the fine subgrain structure leads to superplasticity. In as-cast structures there remains a strong temperature dependence of the flow stress.
6. The strengthening effect of Al_3Sc is accounted for by existing theories of hardening by coherent, ordered precipitates.

Acknowledgement

This work has been supported by the ARC small grants scheme and the Monash development fund.

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Received 29 July 1993
and accepted 19 January 1994