Natural and artificial aging response of semisolid metal processed Al–Si–Mg alloy A356

H. Möller*[1], G. Govender[1] and W. E. Stumpf[2]

International standards for aluminium alloys often permit significant fluctuations in the content of alloying elements. This allows metal suppliers more freedom in preparing these alloys. It is shown that the magnesium content of semisolid metal processed Al–Si–Mg alloy A356 has a significant influence on the natural and artificial aging behaviour of the alloy. Furthermore, natural aging before artificial aging causes the time to peak hardness (T6) to be longer compared to the time when only artificial aging is used. The optimum quality index in this study was obtained using a short solution heat treatment of 1 h at 540°C, no natural aging and artificial aging at 180°C for 1 h. An increase in the magnesium content of the alloy resulted in an increase in the quality index for all the T6 heat treatment cycles studied.

Keywords: Semisolid metal forming, Alloy A356, Natural aging, Artificial aging

Introduction

Semisolid metal (SSM) processing is an effective near net shape manufacturing method in which the metal is formed in the semisolid state. With SSM processing, a semisolid structure free of dendrites with the solid present in a near spherical form is obtained.[1,2] This semisolid mixture flows homogeneously, behaving as a thixotropic fluid with viscosity depending on the shear rate and fraction of solid.[3] With the process of thixocasting, a specially prepared billet of solid material with a globular microstructure is reheated into the semisolid range, followed by a forming process such as high pressure die casting (HPDC). The alternative process of rheocasting involves preparation of an SSM slurry directly from the liquid, followed by HPDC. The higher costs associated with thixocasting have resulted in rheocasting becoming the preferred semisolid process.[2] With conventional liquid HPDC, turbulent die filling is responsible for oxide entrapment, porosity and blistering problems during heat treatment. However, the laminar flow during the die fill for SSM processing avoids the problems of oxide and gas entrapment and also reduces the shrinkage problems with solidification.[4]

The conventional casting alloy A356 is probably the most popular alloy used for semisolid metal forming. This is due to its high fluidity and good 'castability'.[5] The chemical composition limits of this alloy are shown in Table 1.[6] International standards for aluminium alloys often permit significant fluctuations in the content of alloying elements. In Al–Mg–Si alloys containing an excess of silicon, the decomposition of the supersaturated solid solution (SSS) is believed to occur as follows[7]

$$\begin{aligned} \mathrm{SSS} &\rightarrow (\mathrm{Mg}+\mathrm{Si})_{\mathrm{clusters}}/\mathrm{GP(I)}_{\mathrm{spherical}} \\ &\rightarrow \beta/\mathrm{GP(II)}_{\mathrm{needles}} \rightarrow \beta'_{\mathrm{rods}} + \mathrm{Si} + \mathrm{others} \\ &\rightarrow \beta_{\mathrm{plates}} + \mathrm{Si} \end{aligned} \quad (1)$$

where GP=Guinier–Preston zones, β is the equilibrium Mg_2Si, and β' and β'' are the metastable precursors of β.

The natural aging response (room temperature aging after solution treatment and quenching) of alloy A356 is considered to be due to (Mg+Si) clusters and GP zones.[7,8] The precipitation hardening that results from natural aging alone produces the useful T4 temper. Nearly maximum stable values are attained in 4–5 days, therefore, the T4 temper of A356 is usually specified as material that was naturally aged for at least 120 h.[9] Peak hardening with artificial aging (i.e. from the T6 temper) results from the precipitation of the metastable and coherent β''.[7] The Mg content of alloy A356 should have a significant effect on its precipitation hardening characteristics, especially considering the relatively wide composition range that is permissible for this alloying element (Table 1). A higher content of Mg should, first, lead to a higher volume fraction of the second phase β, and most likely also of β' and β'', but second, may even alter the thermodynamic and compositional characteristics of the precipitates by increasing the driving force for nucleation and thus providing a differently sized distribution. Silicon has the strongest influence on the ratio of solid to liquid fraction of all alloying elements in A356 (much stronger than magnesium).[10] A fluctuation of 1 wt-% silicon in aluminium (as is allowed according

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to Table 1) results in a solid fraction change of almost 9% if the casting temperature is held constant.

The heat treatment cycles that are currently applied to semisolid processed components are mostly those that are in use for conventional dendritic casting alloys.[11,12] These heat treatments are not necessarily the optimum treatments for rheocast alloys, as the difference in solidification history and microstructure of rheocast components should be considered. In the casting industry, it is often specified that a dendritic A356 component should be solution treated for 6 h at 540°C.[13] However, Emadi and co-workers[14] have suggested that the optimal solution treatment for dendritic A356 is only 4 h at 540°C. Only limited work has been performed on the optimisation of the solution heat treatment of the SSM processed alloy A356. According to Rosso and Actis Grande,[12] a solution heat treatment of 1 h at 540°C is sufficient to obtain a high level of mechanical properties in the T6 temper (hardness, yield strength, ultimate tensile strength and % elongation). A solution treatment of only 30 min caused the presence of brittle intermetallic phases due to an incomplete solution process. However, according to Dewhirst,[11] the optimum solution treatment time at 540°C is 4 h for SSM processed A356. The effects of natural aging have not received much attention for rheocast alloy A356. For example, Dewhirst[11] varied the natural aging time of semisolid processed A356 between 8 and 24 h before subsequent artificial aging. It was found that increasing the natural aging time beyond 8 h had a slightly negative effect on the tensile properties of the material. Ultimately, it was concluded that artificial aging temperature and time were of greater importance than the previous natural aging time of 8–24 h. The natural aging time that was employed by Rosso and Actis Grande[12] was not documented. A natural aging time of 8 h is frequently used for dendritic A356 components to ensure process uniformity.[11] Finally, the most popular artificial aging treatment for alloy A356 seems to be 170°C for 6 h.[9,12,15] However, the optimum artificial aging treatment for SSM processed A356 was determined to be 4 h at 180°C by both Dewhirst[11] and Rosso and Actis Grande.[12] The objective of this study was to determine the influence of Mg content (at almost constant Si concentration) of rheocast alloy A356 on its aging behaviour (both natural and artificial) and to determine optimum heat treatment conditions.

The quality index (QI) was used in this work to allow comparison of different heat treatment cycles, as well as to investigate the influence of fluctuations in chemical composition. The QI relates the ductility and strength (ultimate tensile strength or UTS) into a single term.[11] It was originally developed by Drouzy *et al.*[16] based on the observation of trends in empirical data. However, Caceres *et al.*[17] have carried out further work to show the fundamental basis of the QI. Shivkumar *et al.*[18] used the QI to optimise the heat treatments of dendritic A356. The QI (specifically for alloy A356) is given by equation (2)[11,16–18]

$$\mathrm{QI\ (MPa)} = \mathrm{UTS\ (MPa)} + 150\log(\%\ \mathrm{elongation}) \qquad (2)$$

The rationale behind this equation lies in the well known phenomenon that for most mechanisms of strengthening in alloys (except for grain refinement), one has to sacrifice some ductility or toughness and a high QI, therefore, aims to find a combination of high strength and high ductility or toughness in the alloy.

Experimental

Semisolid metal slurries of alloy A356 containing different Mg contents (chemical composition given in Table 1) were prepared using the CSIR rheocasting process.[19] Plates ($4 \times 80 \times 100$ mm) were cast in steel moulds with a 50 ton HPDC machine. It can be seen from Table 1 that the Sr content of the three alloys also differs (ranging from 210 to 380 ppm). The amounts of strontium required for modification ranges from 150 to 200 ppm for hypoeutectic castings.[20] However, one of the advantages of using strontium rather than sodium is that overmodification is not believed to cause any significant problems.[20] Solution heat treatment was performed at 540°C for 1 h, followed by a water quench (20°C). The samples were then naturally aged for either 0 h (artificial aging only), 20 h (before reaching the stable T4 temper) or 120 h (stable T4 temper), before being artificially aged at 160 and 180°C to determine artificial aging curves. Vickers hardness numbers (VHN) were determined (using a 20 kg load) from the average of at least four readings per sample. The average hardness values were found to be reproducible within ± 3 VHN for all heat treatment conditions tested. All samples used for microscopy were etched in 0·5%HF solution.

The tensile properties of selected samples were also determined and different heat treatment cycles were compared using the quality index (equation (2)). The tensile samples (substandard size) were machined from the plates (*see* Fig. 1 for the dimensions of the samples). A total of five tensile tests were used for each heat treatment condition.

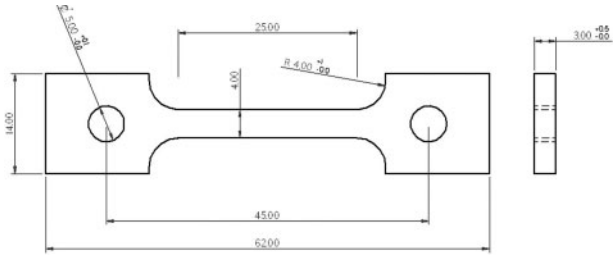
Results and discussion

Optical microscopy

Figure 2 shows a typical optical micrograph of the as cast A356 (with 0·28 wt-%Mg). It is seen that the material has a globular primary grain structure and a fine eutectic. The microstructures for the higher content of Mg alloys are similar to that in Fig. 2. Solution

Table 1 Chemical composition limits (wt-%) for alloy A356[6] and compositions used in this study

	Si	Mg	Fe	Cu	Mn	Zn	Ti	Other (each)	Other (tot)
Min.	6·5	0·25	–	–	–	–	–	–	–
Max.	7·5	0·45	0·20	0·20	0·10	0·10	0·20	0·05	0·15
This study									
	Si	Mg	Fe	Cu	Mn	Zn	Ti	Sr	
Low Mg	7·21	0·28	0·13	0·01	0·01	0·01	0·12	0·038	
Medium Mg	7·14	0·34	0·14	0·01	0·01	0·03	0·08	0·030	
High Mg	7·25	0·45	0·13	0·01	0·01	0·01	0·14	0·021	



1 Dimensions of samples (in mm) used for tensile testing

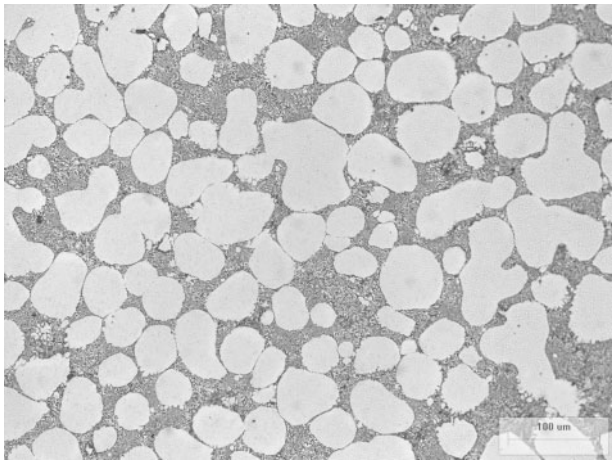
treatment at 540°C for 1 h resulted in the eutectic structure changing to a globular type structure (Fig. 3). The size and shape of the silicon particles were modified with additions of strontium in the A356 used in this work (Table 1). Modified alloys are known to undergo fast spheroidisation, while complete spheroidisation is not achieved in unmodified alloys, even after long solution treatment times.[6,8] Furthermore, microsegregation of silicon and magnesium is not severe in Al–Si–Mg casting alloys and therefore, it only takes a relatively short time to homogenise the alloy and to place the strengthening phase Mg_2Si into solution.[6,8,12]

Natural aging response

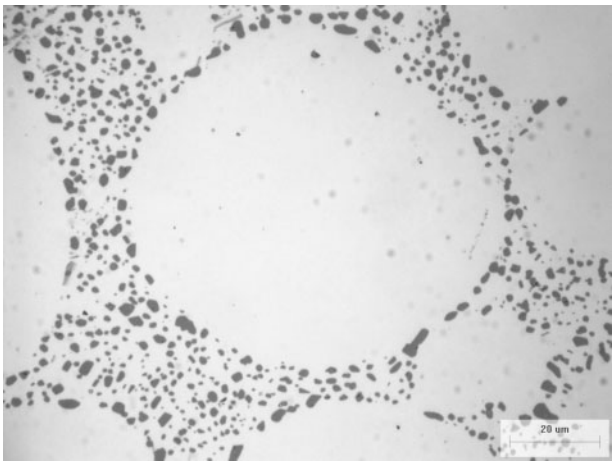
Figure 4 shows the natural aging curve for SSM HPDC alloy A356 after solution treatment at 540°C for 1 h, followed by a water quench. It is seen that the magnesium concentration has a significant effect on the natural aging behaviour of alloy A356. The material is soft directly after quenching (VHN≈53), but thereafter, the hardness increases rapidly. This is likely to be due to the high supersaturation of vacancies that was retained after quenching, which results in the formation of solute clusters and GP zones.[6–8] The hardness levels out at approximately 72, 77 and 85 VHN for magnesium concentrations of 0·28, 0·34 and 0·45% respectively. A linear relationship (with $R^2>0{\cdot}99$) exists between the maximum T4 hardness obtained and the content of Mg of the A356 for the range tested.

Artificial aging at 180°C

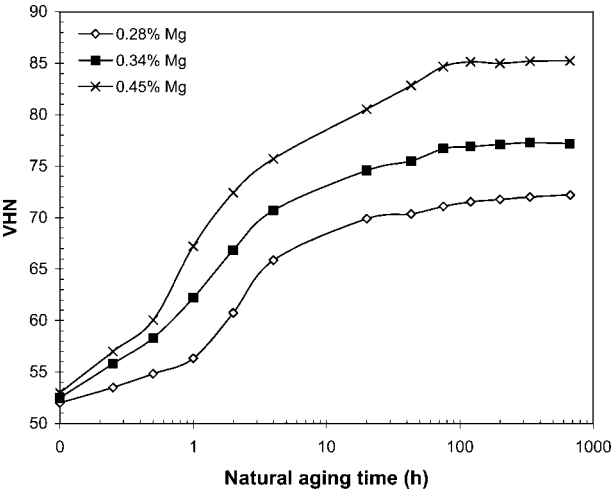
Figure 5 shows artificial aging curves that were determined for alloy A356 after solution treatment at 540°C for 1 h, water quenching and no natural aging. The artificial aging response is very rapid when no natural aging is applied (the converse is true when natural aging



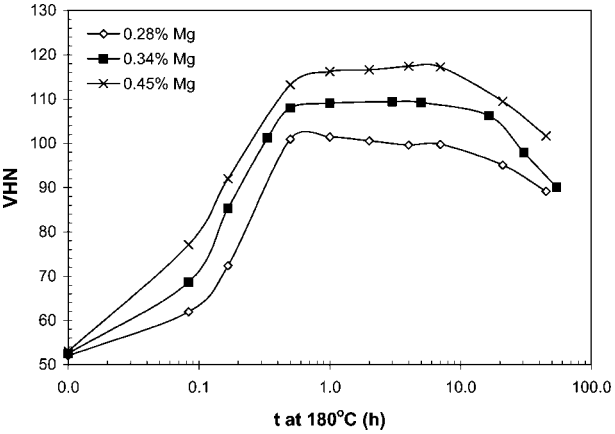
2 Optical micrograph of as cast alloy A356 (with 0·28 wt-%Mg)



3 Optical micrograph of alloy A356 (0·28 wt-%Mg) after solution treatment at 540°C for 1 h

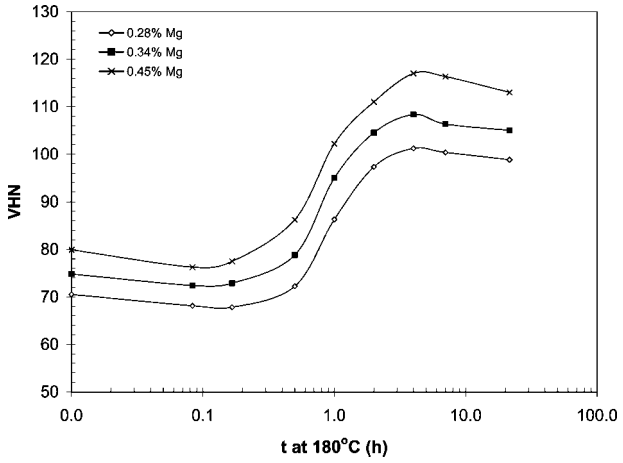


4 Natural aging curves for SSM HPDC A356 following solution treatment at 540°C for 1 h and water quench

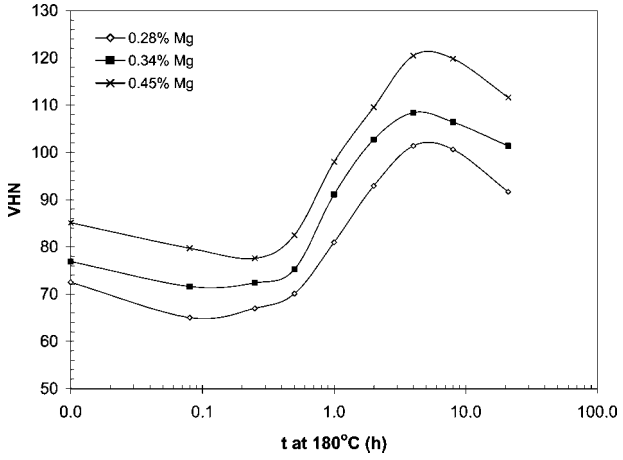


5 Artificial aging curves at 180°C for alloy A356: 0 h natural aging time

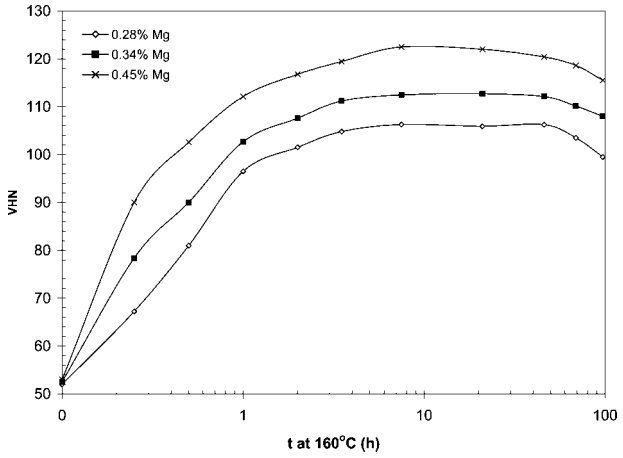
is applied first, as will be shown later). Artificial aging curves were determined for alloy A356 after solution treatment at 540°C for 1 h, water quenching and 20 h natural aging (Fig. 6) or alternatively 120 h natural aging (Fig. 7) followed by artificial aging. The artificial aging response is sluggish when previous natural aging has been employed. This has been attributed to solute clustering during natural aging, and the subsequent



6 Artificial aging curves at 180°C for alloy A356: 20 h natural aging time

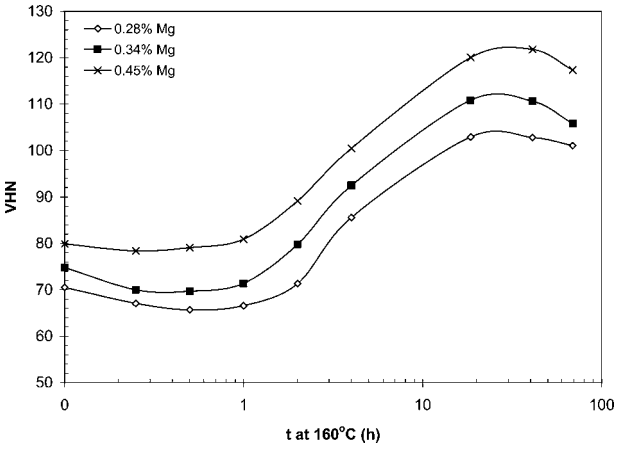


7 Artificial aging curves at 180°C for alloy A356: 120 h natural aging time



8 Artificial aging curves at 160°C for alloy A356: 0 h natural aging time

dissolution of these clusters initially before artificial aging can commence.[7,8] It is the reversion of these clusters which most likely results in the loss in hardness during the initial stages of artificial aging (Figs. 6 and 7). It is seen that the extent of the loss is recovered (presumably by the precipitation of β'' particles) upon further aging. However, the initial dissolution of the clusters and GP zones causes the time to peak hardness



9 Artificial aging curves at 160°C for alloy A356: 20 h natural aging time

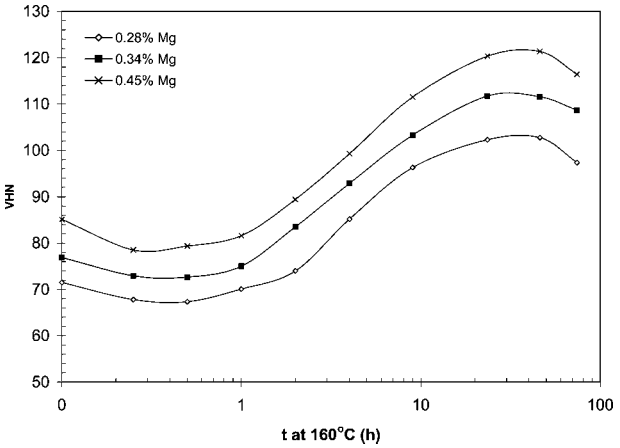
(T6) to be longer compared to the case when no natural aging is used. The increase in natural aging from 20 to 120 h does not significantly influence the subsequent artificial aging response. Dewhirst[11] also found no major differences in the artificial aging response by increasing the prior natural aging time from 8 to 24 h.

Artificial aging at 160°C

Artificial aging curves were also determined at 160°C after no natural aging (Fig. 8), after 20 h natural aging (Fig. 9) and after 120 h natural aging (Fig. 10). As expected, the maximum hardness values achieved are slightly higher than at 180°C, due to the lower solubility of strengthening phases at lower temperatures, but likely also due to a higher thermodynamic driving force for nucleation of the second phases, leading to a finer size distribution. Unfortunately, due to the lower diffusion rates at 160°C, the time to peak hardness is reached after much longer times than with artificial aging at 180°C (Table 2). It can be seen from Table 2 that it takes approximately six times longer to reach peak hardness at 160°C compared to at 180°C. The diffusivity of silicon in aluminium is about double that of magnesium in aluminium at the studied temperatures of 160 and 180°C.[21] Using the diffusion data of magnesium in aluminium published by Du and co-workers[21] ($D_o = 1{\cdot}5 \times 10^{-5}$ m^2 s^{-1}, $Q = 121$ kJ mol^{-1}), it can be calculated that the diffusivity at 180°C is approximately five times higher than at 160°C. This corresponds reasonably well with the increase in time to peak hardness observed (Table 2).

Table 2 Time to peak hardness as function of the natural aging time and artificial aging temperature

Artificial aging temperature, °C	Time to peak hardness, h
No natural aging before artificial aging	
160	6
180	1
20 h natural aging before artificial aging	
160	25
180	4
120 h natural aging before artificial aging	
160	30
180	5



10 Artificial aging curves at 160°C for alloy A356: 120 h natural aging time

The linear relationship (with $R^2>0{\cdot}99$) between content of Mg and peak hardness also holds with all T6 heat treatment cycles. Again, the increase in natural aging time from 20 to 120 h does not have a significant influence on the subsequent artificial aging behaviour of this alloy.

Tensile tests

T6 heat treatments were performed on samples for tensile tests based on the artificial aging curves presented in Figs. 5 and 6. The popular 'traditional' T6 heat treatment[9,15] of solution treatment for 6 h at 540°C, water quenching, natural aging for 20 h and artificial aging for 6 h at 170°C was also used for comparison. It was decided not to do artificial aging at 160°C, due to the relatively long times needed to reach peak hardness (Table 2), without reaching significantly higher hardnesses compared to 180°C artificial aging. Details of the T6 heat treatment cycles used for tensile testing are shown in Table 3. The time needed for each heat treatment is also specified in Table 3. It is known that the production time and cost of the T6 heat treatment is considerable.[12,13] Shortening the total time of the T6 heat treatment cycle would have an important impact on manufacturing cost and productivity. The results of the tensile tests are shown in Table 4. From this table, it is seen that, for all three compositions, the best QI is achieved using the '540-1, 0-NA, 180-1' heat treatment, which, advantageously, is also the shortest heat treatment (Table 3). Application of this very short heat treatment cycle would need precise temperature control and reliable heat treatment furnaces. The QI of the '540-1, 20-NA, 180-4' heat treatment falls within the experimental standard deviation and the difference compared to the 'traditional' heat treatment may not be significant for the 0·34 and 0·45%Mg samples. However, the '540-1, 20-NA, 180-4' heat treatment is still much shorter than the 'traditional' T6 heat treatment. This heat treatment can rather be used when it is not possible to artificially age the A356 immediately after the solution heat treatment and quench. In such a case, it would probably be more advantageous to use a natural aging time of 8 h (rather than 20 h) to shorten the T6 heat treatment cycle and to ensure process uniformity.[11] It has also been claimed that an advantage of prior natural aging (6–20 h) is to reduce scatter in properties.[14] However, this claim is not supported by the standard deviations that were obtained in this study (Table 4).

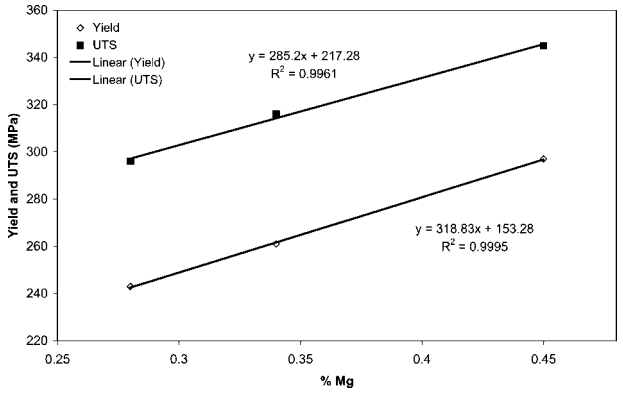
It is also evident from Table 4 that the QI increases when the magnesium content is increased for each heat treatment. This is due to the increase in strength of alloy A356, without significantly influencing the elongation, when the content of Mg is increased (Table 4). This phenomenon will be studied further by employing transmission electron microscopy. It needs to be determined whether the content of Mg only influences the volume fraction of β, β' and β'', or if it might cause a fundamental change in the precipitation behaviour of this alloy. Figures 11–13 show the yield strength and UTS as a function of the content of Mg for the three different heat treatments. The excellent correlation that exists in all cases makes it possible to interpolate the

Table 3 T6 Heat treatment cycles used for tensile testing

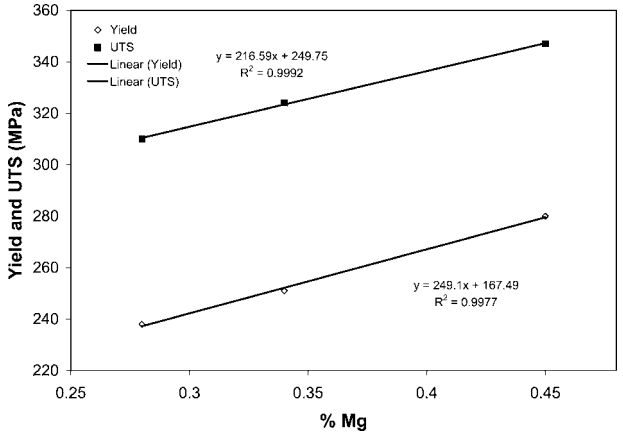
Heat treatment	Solution treatment	Natural aging	Artificial aging	Total furnace time, h	Total time, h
540-6, 20NA, 170-6	540°C for 6 h	20 h	170°C for 6 h	12	32
540-1, 0NA, 180-1	540°C for 1 h	0 h	180°C for 1 h	2	2
540-1, 20NA, 180-4	540°C for 1 h	20 h	180°C for 4 h	5	25

Table 4 Yield strength, UTS, % elongation and QI of heat treated samples: standard deviations from five values for tensile properties are indicated in brackets

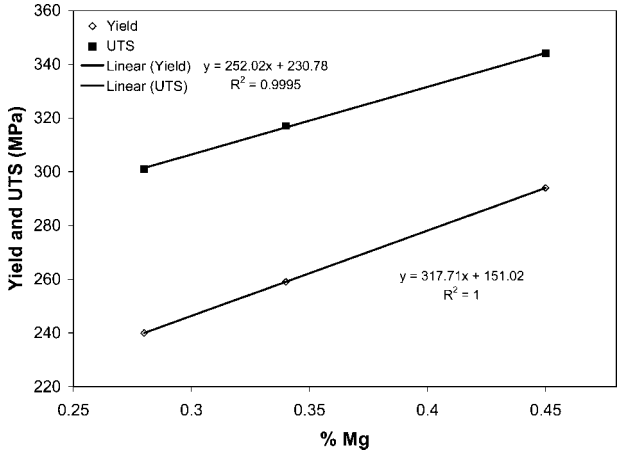
Heat treatment	Yield strength, MPa	UTS, MPa	% elongation	QI, MPa
0·28%Mg				
540-6, 20NA, 170-6	243 (4·1)	296 (6·5)	7·2 (1·3)	425
540-1, 0NA, 180-1	238 (7·2)	310 (8·9)	10·4 (1·2)	463
540-1, 20NA, 180-4	240 (2·8)	301 (3·1)	9·4 (0·9)	447
0·34%Mg				
540-6, 20NA, 170-6	261 (4·5)	316 (5·9)	8·3 (2·1)	454
540-1, 0NA, 180-1	251 (3·8)	324 (7·3)	10·1 (1·5)	475
540-1, 20NA, 180-4	259 (5·2)	317 (7·1)	8·8 (1·4)	459
0·45%Mg				
540-6, 20NA, 170-6	297 (5·0)	345 (5·6)	7·3 (2·3)	474
540-1, 0NA, 180-1	280 (6·0)	347 (3·6)	9·8 (1·4)	496
540-1, 20NA, 180-4	294 (5·8)	344 (3·1)	7·1 (1·4)	472



11 Yield strength and UTS of alloy A356 as function of Mg content (wt-%) of alloy for 'traditional' T6 heat treatment (540-6, 20NA, 170-6)



12 Yield strength and UTS of alloy A356 as function of Mg content (wt-%) of alloy for '540-1, 0NA, 180-1' T6 heat treatment



13 Yield strength and UTS of alloy A356 as function of Mg content (wt-%) of alloy for '540-1, 20NA, 180-4' T6 heat treatment

curves if yield strength and UTS at other magnesium contents are to be estimated.

Conclusions

The conclusions of this study are:

1. The magnesium content of alloy A356 has a significant influence on the natural and artificial aging behaviour of the alloy.

2. The dissolution of solute clusters and GP zones (which are formed during natural aging) during the initial stages of artificial aging causes softening of the alloy. This in turn causes the time to peak hardness (T6) to be longer compared to the time when no natural aging is used for all Mg contents studied.

3. Artificial aging at 160°C produces slightly higher peak hardness compared to artificial aging at 180°C, but the time to peak hardness is significantly increased.

4. The quality index constitutes a useful tool to gauge the effect of changes to the heat treatment cycles of SSM HPDC A356. The best QI in this study was obtained using a short solution heat treatment of only 1 h at 540°C, no natural aging and artificial aging at 180°C for 1 h.

5. An increase in the magnesium content results in an increase in the QI for all the heat treatments studied.

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References

1. M. C. Flemings and R. A. Martinez: *Solid State Phenom.*, 2006, **116–117**, 1–8.
2. H. V. Atkinson: *Prog. Mater. Sci.*, 2005, **50**, 341–412.
3. O. Lashkari and R. Ghomashchi: *J. Mater. Process. Technol.*, 2007, **182**, 229–240.
4. W. L. Winterbottom: *Metall. Sci. Technol.*, 2000, **18**, 5–10.
5. D. Liu, H. V. Atkinson, P. Kapranos, W. Jirattiticharoean and H. Jones: *Mater. Sci. Eng. A*, 2003, **A361**, 213–224.
6. J. R. Davis: 'ASM specialty handbook: aluminium and aluminium alloys', 315–316, 718; 1993, Materials Park, OH, ASM International.
7. A. K. Gupta, D. J. Lloyd and S. A. Court: *Mater. Sci. Eng. A*, 2001, **A301**, 140–146.
8. H. Möller, G. Govender and W. E. Stumpf: *Open Materials Science Journal*, 2008, **2**, 6–10.
9. G. Govender, L. Ivanchev, N. Jahajeeah and R. Bëan: *Solid State Phenom.*, 2006, **116–117**, 501–504.
10. H. Kaufmann, W. Fragner, U. Galovsky and P. J. Uggowitzer: Proc. 2nd Int. Light Metals Technology Conf., St. Wolfgang, Austria, June 2005, ARC Leichtmetallkompetenzzentrum Ranshofen GmbHm, 169–177.
11. B. A. Dewhirst: 'Optimization of the heat treatment of semi solid processed A356 aluminum alloy', MSc thesis, Worcester Polytechnic Institute, Worcester, MA, USA, 2005.
12. M. Rosso and M. Actis Grande: *Solid State Phenom.*, 2006, **116–117**, 505–508.
13. D. L. Zhang, L. H. Zheng and D. H. StJohn: *J. Light Met.*, 2002, **2**, 27–36.
14. D. Emadi, L. V. Whiting, M. Sahoo, J. H. Sokolowski, P. Burke and M. Hart: Proc. 132th TMS Annual Conf., San Diego, CA, USA, March 2003, TMS, 983–989.
15. M. Badiali, C. J. Davidson, J. R. Griffiths and A. Zanada: Proc. 6th Int. Conf. on 'Semi-solid processing of alloys and composites', Turin, Italy, September 2000, Edimet, 349–354.
16. M. Drouzy, S. Jacob and M. Richard: *AFS Int. Cast Met. Res. J.*, 1980, **5**, 43–50.
17. C. H. Caceres, M. Makhlouf, D. Apelian and L. Wang: *J. Light Met.*, 2001, **1**, 51–59.
18. S. Shivkumar, S. Ricci, B. Steenhoff, D. Apelian and G. Sigworth: *AFS Trans.*, 1989, **138**, 791–810.
19. L. Ivanchev, D. Wilkins and G. Govender: Proc. 8th Int. Conf. on 'Semi-solid processing of alloys and composites', Limassol, Cyprus, September 2004, WPI's Metal Processing Institute, Paper 152.

20. I. J. Polmear: 'Light alloys from traditional alloys to nanocrystals', 4th edn, 220; 2006, Amsterdam, Elsevier.

21. Y. Du, Y. A. Chang, B. Huang, W. Gong, Z. Jin, H. Xu, Z. Yuan, Y. Liu, Y. He and F. Xie: *Mater. Sci. Eng. A*, 2003, **A363**, 140–51.