The influence of natural ageing on the artificial ageing response of Al-Si-Cu-Mg casting alloys

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The T6 heat treatment is commonly used to increase the strength of gravity cast Al-Si components containing Cu and/or Mg. The artificial ageing response is known to be affected by the thermal history, such as solution treatment, quench rate, natural ageing and heating rate to the artificial ageing temperature. The influence of natural ageing on the artificial ageing response was investigated for three alloys; Al-8Si-0.4Mg, Al-7Si-3Cu and Al-8Si-3Cu-0.4Mg. Natural ageing had a strong influence on the ageing response of the Al-Si-Mg alloy in the underaged condition and the strength increase was strongly reduced. Despite this, the time to peak yield strength as well as its magnitude were not strongly affected by natural ageing. No clear influence of natural ageing was observed for the Al-Si-Cu alloy. For the Al-Si-Cu-Mg alloy the ageing response seems to depend on the natural ageing time. Natural ageing of 3 weeks shifted the peak yield strength to shorter ageing times and its magnitude was decreased a little compared to direct ageing after quench. Natural ageing of 1 day gave the least beneficial properties after artificial ageing.

INTRODUCTION

Natural ageing is known to have an influence on mechanical properties obtained after artificial ageing. A substantial number of experiments have been performed on wrought Al-Mg-Si alloys due to the negative influence of natural ageing observed for short artificial ageing times, i.e. for paint-bake treatments. The negative influence is largest for short artificial ageing times, while the strength at peak aged condition is almost recovered [1-4].

Clusters of solute atoms form in the matrix during natural ageing consuming both solute atoms and vacancies. The clusters, which form in Al-Mg-Si alloys, can not act as nuclei for β'' precipitates [5-7]. There are however different opinions regarding the stability of the clusters when heated to the artificial ageing temperature. According to Pashley et al. [8] the stability of the clusters is determined by their radius, the supersaturation of the matrix and the ageing temperature. When the clusters have a too small radius to be stable at the artificial ageing temperature they will start to dissolve [8]. When they dissolve solute is released to the matrix and the supersaturation increases and some of the clusters may become stable or new clusters may nucleate [8]. The theory of clusters dissolving during artificial ageing is strengthened by the observation of a decrease in yield strength at initial artificial ageing and the occurrence of a dissolution peak on the differential scanning calorimetry trace for naturally aged samples [9]. There are however indications in the literature that the size of the clusters is not the main parameter determining the stability, but rather the nature of the clusters [6, 7, 10]. According to Yamada et al. [7] stable Si-rich clusters are formed during natural ageing which will neither nucleate the β'' phase nor dissolve at artificial ageing.

Regardless the stability of the clusters, a lower supersaturation of solute atoms and vacancies will be available for formation of new clusters during artificial ageing, which explains the observations of microstructures having a lower number density of coarser β'' precipitates for all artificial ageing times when natural ageing is applied for Al-Mg-Si wrought alloys [3-6, 11].

The influence of natural ageing on the artificial ageing response of Al-Si casting alloys is a less investigated field. A number of studies of Al-Si-Mg alloys have been reported [12-17]. Möller et al. [14] have studied the influence of natural ageing on the artificial ageing response for Al-7Si-Mg alloys having various Mg concentrations. Natural ageing times of 0 h, 20 h and 120 h and artificial ageing temperatures of 160°C and 180°C were used. In Fig. 1 natural ageing is seen to have a large influence on the hardness during underageing. The natural aged samples have a higher hardness prior to artificial ageing due to formation of clusters at room temperature. The hardness decreases at initial artificial ageing indicating that the clusters formed during natural ageing dissolve. In Fig. 1 it is also observed that the peak hardness is independent of natural ageing. Sha et al. [18] studied the precipitates forming in an Al-7Si-0.5Mg alloy aged at 180°C with and without natural ageing using transmission electron microscopy and atom probe tomography. In agreement with wrought Al-Mg-Si alloys the β'' precipitates formed when artificial ageing is applied directly after quench have a higher number density and are finer compared to the precipitates formed when natural ageing is used.

Despite the difference in number density and size of the precipitates similar peak hardness is obtained.

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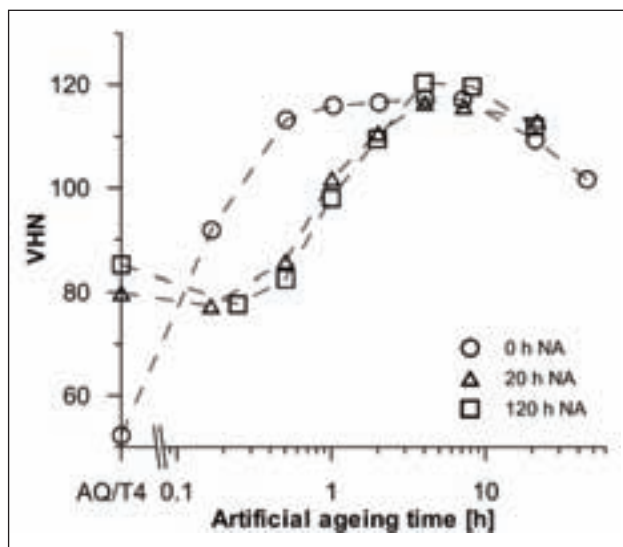High Tech DieCasting, Vicenza, 9-11 February 2012*

FIG. 1 *Influence of natural ageing on hardness after artificial ageing at 180°C for an Al-7Si-0.45Mg alloy [14].*

Influenza dell'invecchiamento naturale sulla durezza dopo invecchiamento artificiale a 180°C per una lega Al-7Si-0.45Mg [14].

The data for Al-Si-Cu-Mg alloys in the literature is poor and diverging. Reif et al. [19] report only a small increase in hardness when natural ageing of 4 h was applied prior to artificial ageing at 160°C for an Al-9Si-3.5Cu-0.5Mg alloy. On the other hand, a large increase in yield strength was observed by Geier et al. [20] for an Al-9Si-2.8Cu-0.3Mg-0.5Zn alloy for natural ageing of 3 days to 3 weeks followed by artificial ageing at 140°C to 240°C. The aim of the study is to investigate the influence of natural ageing on the artificial ageing response for three different Al-Si casting alloys. First, the investigation for Al-Si-Mg alloys made by Möller et al. [14] is repeated. Their study is the only one showing the influence of natural ageing on the whole ageing curve and the results need to be confirmed. Secondly, the Al-Si-Cu and Al-Si-Cu-Mg alloys are studied as there is a lack of data for these alloys. For the Al-Si-Cu-Mg alloy there is an interest to see if natural ageing influences the fraction of β'' and Q'' precipitates formed after artificial ageing, and thereby the shape of the ageing curves, which has been observed for wrought aluminium alloys [21].

MATERIAL AND EXPERIMENTS

Three Al-Si alloys were cast having various Mg and Cu concentrations, as shown in Table 1. Sr modification was used. Cylindrical rods (length 18 cm, diameter 1 cm) were cast in a preheated permanent mould. The rods were recast with the gradient solidification technique which gives samples with a low content of porosity defects thanks to the good feeding. The gradient solidification technique allows the solidification rate and hence the coarseness of the microstructure to be chosen. Rods with secondary dendrite arm spacings (SDAS) of approximately 25 μm were produced, which corresponds to a coarseness of the

microstructure obtained for gravity die casting. Tensile test bars with a gauge length of 50 mm and a diameter of 7 mm were machined from the rods prior to heat treatment.

Solution treatment was conducted in an electrical furnace at a temperature of 495°C for the Cu containing alloys and at 530°C for the Al-Si-Mg alloy. A time of 3 h was chosen as earlier wavelength dispersive spectroscopy measurements on similar alloys has shown that 3 h is enough to dissolve particles which are not stable at the solution treatment temperature to obtain a homogenous and high concentration of alloying elements in solid solution [22]. The samples were quenched in 50°C water. Three different natural ageing times prior to artificial ageing were used: 0 min, 1 day and 3 weeks. Artificial ageing was conducted in a forced circulation air furnace at 170°C or 210°C for various times between only heating up and 250 h. The times needed for heating the samples to the solution treatment temperature (10-15 min) and the artificial ageing temperature (20 min) are excluded from the recorded times. Tensile tests were performed at a constant strain rate of 0.5 mm/min using a Zwick/Roell Z100 machine equipped with a 100 kN load cell and a clip-on 20 mm gauge length extensometer.

Two tensile test bars was used for each ageing condition.

RESULTS AND DISCUSSION

In Fig. 2 natural ageing is seen to have a large influence on the ageing curves of the Al-Si-Mg alloy for 170°C and 210°C. The results for ageing at 170°C is in agreement with the results presented by Möller et al. [14] in Fig. 1. A low agehardening response is observed for short ageing times for natural aged samples while the peak yield strength is independent of natural ageing. The shape of the ageing curve of the natural aged sam-
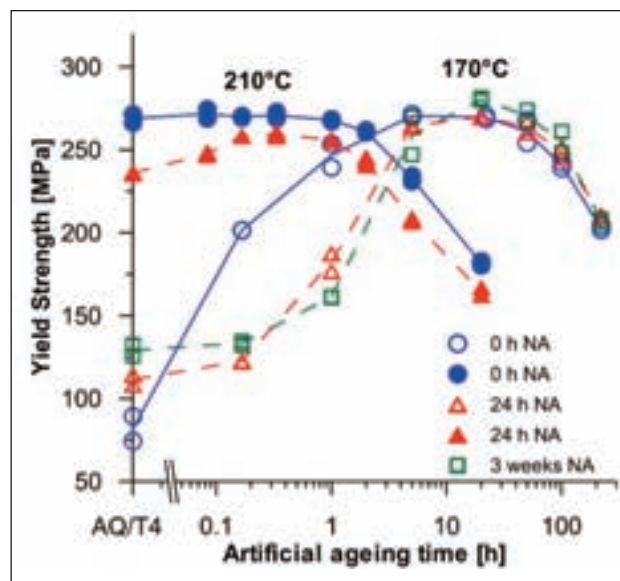

FIG. 2 *Artificial ageing curves for 170°C and 210°C for the Al-Si-Mg alloy naturally aged various times.*

Curve di invecchiamento artificiale a 170°C e 210°C per la lega Al-Si-Mg invecchiata naturalmente per tempi diversi.

TABLE 1
Alloy composition in wt%.

Composizione delle leghe, in peso%.

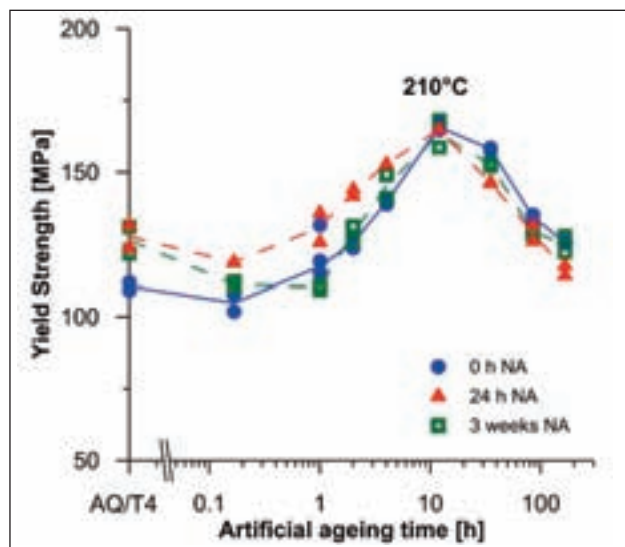|	Si	Cu	Mg	Fe	Al	Sr (ppm)
Al-Si-Mg	7.8	0.0	0.40	0.09	Bal.	391
Al-Si-Cu	7.2	3.2	0.00	0.09	Bal.	193
Al-Si-Cu-Mg	7.8	3.4	0.40	0.08	Bal.	366

FIG. 3 *Artificial ageing curves for 210°C for the Al-Si-Cu alloy naturally aged various times.*

Curve di invecchiamento artificiale a 210°C per la lega Al-Si-Cu invecchiata naturalmente per tempi diversi.
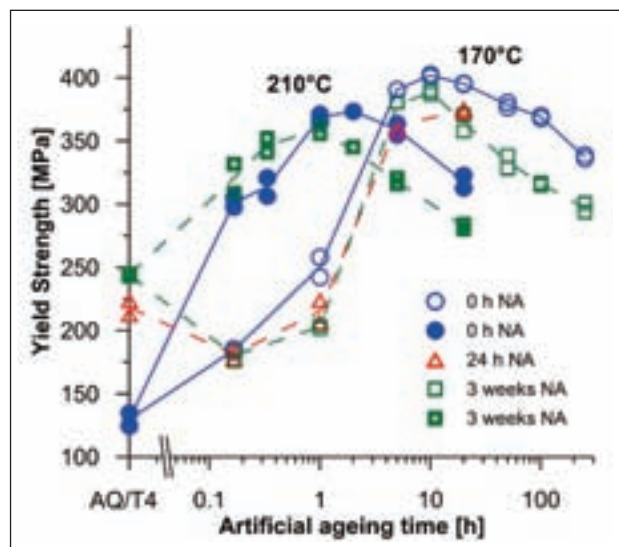

FIG. 4 *Artificial ageing curves for 170°C and 210°C for the Al-Si-Cu-Mg alloy naturally aged various times.*

Curve di invecchiamento artificiale a 170°C e 210°C per la lega Al-Si-Cu-Mg invecchiata naturalmente per tempi diversi.

ples do not show the plateau seen for directly aged samples and the time to peak is shifted towards longer ageing times. The ageing curves after natural ageing of 24 h and 3 weeks are similar, meaning that most of the cluster formation takes place during the first 24 h of natural ageing. The higher yield strength after 3 weeks of natural ageing compared to 24 h however indicates that some cluster growth takes place even after 24 h. The influence of natural ageing on artificial ageing at 210°C seems to be slightly different compared to ageing at 170°C, with a peak yield strength about 10 MPa lower compared to the directly aged samples. The high yield strength observed for ageing at 210°C after 0 min is due to precipitation taking place during the 20 min long heating up time.

Artificial ageing was only done at 210°C for the Al-Si-Cu alloy as the agehardening response is very slow at 170°C and the peak yield strength is reached after 170 h or more [23]. Natural ageing of the Al-Si-Cu alloy does not have a clear influence of the ageing response at 210°C as seen in Fig. 3. The peak hardness, about 165 MPa, and the time to peak, 12 h, are not influenced by natural ageing. The increase in yield strength during underageing shows some differences, where samples natural aged 24 h show higher strengths. It is unclear if this effect is real, or a result of differences in the experimental method. The ageing curves for 0 h and 3 weeks of natural ageing were done by one person, while the ageing curve for 24 h was done later by another person. To check the influence of the experimental method, one sample for ageing directly after quench for 1 h was repeated and the result was close to that obtained for natural ageing of 24 h before ageing, indicating that there is no real influence of natural ageing of Al-Si-Cu alloys on the artificial ageing response. Finally, natural ageing of 24 h and 3 weeks results in similar hardness, meaning that no further growth of clusters takes place after 24 h at room temperature. This is a further indication that the ageing curves for samples naturally aged 24 h and 3 weeks should be similar.

The influence of natural ageing on the artificial ageing response for the Al-Si-Cu-Mg alloy is shown in Fig. 4. A large increase in strength is obtained during the first 24 h of natural ageing and the strength continues to increase although at a slower rate up

to 3 weeks .The highest yield strength is obtained with artificial ageing directly after quench for both ageing temperatures, 402 MPa at 170°C and 374 MPA at 210°C . Natural ageing of 3 weeks gives slightly lower peak strength, while natural ageing of 24 h gives a further decrease in peak strength. To complete the series of ageing curves and make the evaluation easier ageing curves for a similar alloy [23], Al-8.5Si-3.1Cu-0.47Mg, naturally aged 24 h prior to artificial ageing was included in Fig. 5. Average values based on two tensile test bars are shown in Fig. 5 to increase the readability of the figure.

Overageing is observed to start earlier for natural ageing of 3 weeks compared to natural ageing of 0 h or 24 h. This seems to be valid for both ageing temperatures. For ageing at 210°C the 3 week naturally aged samples give higher yield strengths for short ageing times, while the opposite behaviour is observed for ageing at 170°C. A clear decrease in yield strength at initial artificial ageing at 170°C is observed for the naturally aged samples (Fig. 5b) indicating that clusters formed at room temperature dissolve. For artificial ageing at 210°C (Fig. 5a) no such decrease in strength is seen. Two interpretations are possible; either dissolution and reformation of clusters have already taken place after 10 min at 210°C, or the clusters formed at room temperature are stable at 210°C. The higher strength for short ageing times for the 3 weeks naturally aged samples compared to the directly aged samples supports the interpretation that the clusters formed at room temperature are stable at 210°C.

One further reason for studying the influence of natural ageing on the artificial ageing response of Al-Si-Cu-Mg alloys is the occurrence of two different types of ageing curves in the literature. Li et al. [24] report ageing curves for Al-Si-Cu-Mg alloys having a time to peak which is comparable to that for Al-Si-Mg alloys, while Wang et al. [25] report ageing curves for Al-Si-Cu-Mg alloys which has a longer time to peak. The β'' phase was reported to be the main strengthening precipitate in samples having the shorter time to peak, while the Q'' phase was reported to be the main precipitate in samples having longer time to peak. The β'' phase has a higher and faster strength contribution compared to the Q'' phase and explains the difference in appearance of the ageing curves. It is however not known what causes

FIG. 5
Artificial ageing curves for a) 210°C and b) 170°C for the Al-Si-Cu-Mg alloy naturally aged various times. Data from [23] for natural ageing of 24 h prior to artificial ageing.
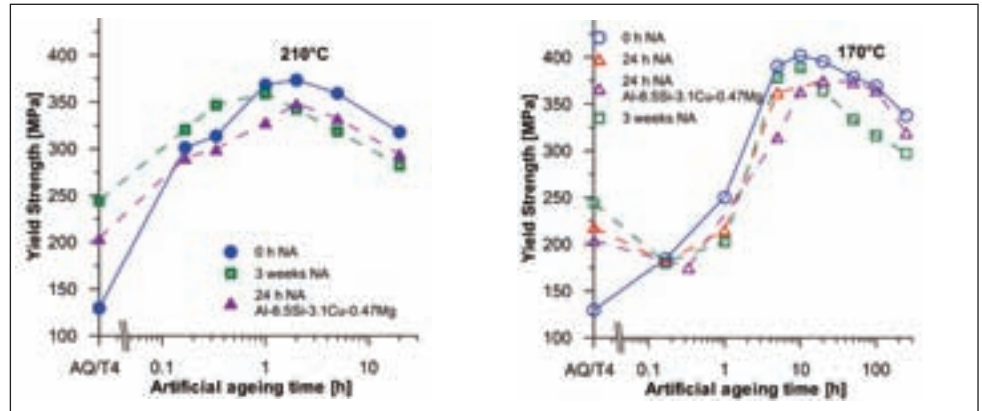
Curve di invecchiamento artificiale alle temperature di: a) 210°C e b) 170°C, per la lega Al-Si-Cu-Mg invecchiata naturalmente per tempi diversi. Dati da [23] per l'invecchiamento naturale di 24 h prima dell'invecchiamento artificiale.

FIG. 6
Comparison of ageing curves for Al-Si-Mg and Al-Si-Cu-Mg alloys. Only the contour of the ageing curves is shown.

Confronto fra le curve di invecchiamento delle leghe Al-Si-Mn e Al-Si-Cu-Mg. Viene mostrata solo la sagoma delle curve di invecchiamento.
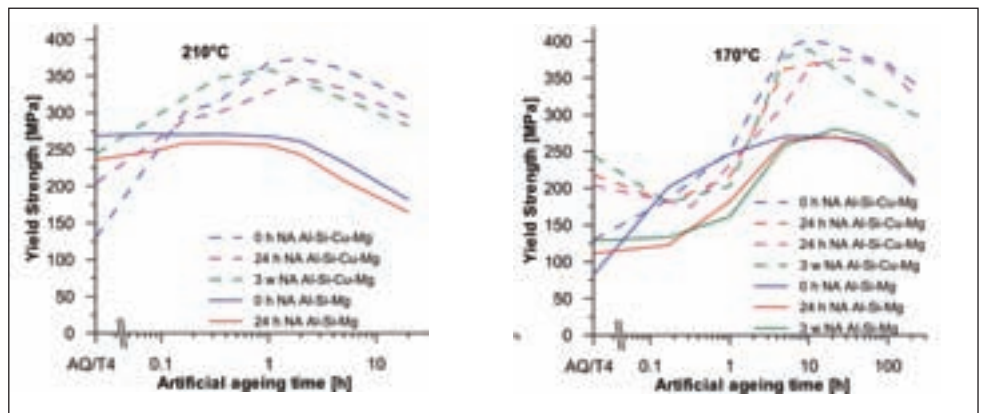
the formation of different precipitates for similar alloys. From wrought alloys it is known that the fraction of the Ω'' phase increases with ageing temperature, ageing time and natural ageing [21, 26, 27]. If the fraction of the Ω'' precipitate increases when natural ageing is used also for Al-Si-Cu-Mg casting alloys a lower peak shifted towards longer ageing times should be observed. A tendency of this behaviour is seen for samples naturally aged for 24 h prior to artificial ageing, while this is not seen for samples naturally aged 3 weeks.

A comparison of the time to peak for Al-Si-Mg and Al-Si-Cu-Mg alloys is made in Fig. 6.

The time to peak for ageing directly after quench at 210°C is much longer for the Al-Si-Cu-Mg alloy (1-2 h) compared to the Al-Si-Mg alloy (only heating to the ageing temperature). A longer time to peak for the Al-Si-Cu-Mg alloy seems to be valid also for natural aged samples. The situation is different for ageing at 170°C. The time to peak is slightly longer for the Al-Si-Cu-Mg alloy (10 h) compared to the Al-Si-Mg alloy (5-22 h) for direct ageing. For natural ageing of 24 h the time to peak is similar for the two alloys (20 h). For natural ageing of 3 weeks the behaviour changes and the Al-Si-Cu-Mg alloy has a shorter time to peak (10 h) compared to the Al-Si-Mg alloy (20 h). Based on the time to peak, the Ω'' phase is indicated to form in the Al-Si-Cu-Mg alloy during ageing at 210°C. For ageing at 170°C the same main precipitate, probably β'', form in the Al-Si-Cu-Mg alloy for ageing after 0 h or 24 h of natural ageing. A higher fraction of Ω'' phase at a higher ageing temperature is in agreement with literature [21]. There are still many uncertainties regarding the influence of natural ageing on the type and fraction of precipitates forming during artificial ageing for the Al-Si-Cu-Mg alloy. This is far too complex to be investigated using only ageing curves and

further studies will be conducted using differential scanning calorimetry.

CONCLUSIONS
- A strong influence of natural ageing on the artificial ageing response for Al-Si-Mg alloys for short ageing times was confirmed.
- No clear influence of natural ageing was observed for the Al-Si-Cu alloy.
- Ageing curves for two ageing temperatures and different natural ageing times have been presented for the Al-Si-Cu-Mg alloy.
- Direct artificial ageing after quench gave the highest yield strength for the Al-Si-Cu-Mg alloy.
- Natural ageing of 3 weeks is preferable to 24 h for the Al-Si-Cu-Mg alloy.
- Natural ageing of 3 weeks resulted in a shorter time to peak at 210°C compared to without natural ageing, while no shift was seen for ageing at 170°C for the Al-Si-Cu-Mg alloy.
- The influence of natural ageing on the artificial ageing response of the Al-Si-Cu-Mg alloy needs to be studied further.

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Abstract

L'influenza dell'invecchiamento naturale sulla risposta all'invecchiamento artificiale nelle leghe per colata Al-Si-Cu-Mg

Parole chiave: trattamenti termici – invecchiamento – alluminio e leghe

Il trattamento termico T6 è comunemente usato per aumentare le caratteristiche meccaniche di componenti di Al-Si contenenti Cu e / o Mg colati per gravità. La risposta all' invecchiamento artificiale è nota per essere influenzata dalla storia termica, quali ad esempio il trattamento di solubilizzazione, la velocità di raffreddamento, il tasso di invecchiamento naturale e la velocità di riscaldamento alla temperatura di invecchiamento artificiale. L'influenza dell' invecchiamento naturale sulla risposta all' invecchiamento artificiale è stato studiata per tre leghe; Al-8Si-0,4 Mg, di Al-7Si-3Cu e Al-8Si-3Cu-0.4Mg.

L'invecchiamento naturale ha avuto una forte influenza sulla risposta all' invecchiamento della lega Al-Si-Mg in condizione di sotto-invecchiamento e l'aumento della resistenza meccanica è stato fortemente ridotto. Nonostante questo, il tempo per raggiungere il picco di snervamento e la sua grandezza non sono stati significativamente influenzati dall'invecchiamento naturale. Per la lega Al-Si-Cu non è stata osservata alcuna netta influenza dell' invecchiamento naturale. Per la lega Al-Si-Cu-Mg la risposta all'invecchiamento sembra dipendere dal tempo di invecchiamento naturale. L'invecchiamento naturale di 3 settimane ha spostato il picco di snervamento verso tempi di invecchiamento più brevi e la sua grandezza è diminuita un poco rispetto a quella da invecchiamento diretto dopo tempra. L'invecchiamento naturale di 1 giorno ha provocato il minor miglioramento delle caratteristiche dopo l'invecchiamento artificiale.