

TALAT Lecture 1601

Process Modelling Applied to Age Hardening Aluminium Alloys

19 pages, 9 figures

(Level Advanced 2)

prepared by Ø. Grong, University of Trondheim, Norway*)

Objectives:

- 1. Description of the methodology for physical modelling of materials problems, with particular emphasis on heat treatment and welding of age hardening alloys materials.
- 2. Establish mathematical relations between different process variables (e.g. alloy composition, heat treatment procedure, welding conditions) and the alloy strength or hardness, based on sound physical principles (e.g. thermodynamics, kinetic theory, dislocation mechanics).
- 3. Motivate faster process development, optimization of process and properties and development of real-time control.

Prerequisites:

Students:

Graduate education in metallurgy, materials science, research experience engineering Trainers:

Research in physical metallurgy, thermodynamics, kinetic theory and dislocation theory.

Date of Issue: 1994

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^{*)} based on a paper given by H.R.Shercliff, Ø.Grong, O.R.Myhr and M.F.Ashby at the 3rd International Conference on Aluminium Alloys; ICAA3 - Aluminium Alloys: Their Physical and Mechanical Properties.

Editors: L.Arnberg, O.Lohne, E.Nes and N.Ryum, SINTEF, Metallurgy, Trondheim, Norway 22-27 June 1992.

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Summary

Process modelling methods have been applied to the thermal treatment of age hardening aluminium alloys. A general methodology for modelling problems of this type is described, based on evolution of internal state variables which describe the microstructure and the subsequent mechanical properties. The approach is first illustrated by a model for isothermal ageing of aluminium alloys. Modelling of processes in which the temperature varies with time is discussed. A second model describes the development of microstructure and strength following reheating of artificially aged (T6) Al-Mg-Si alloys, in particular by welding. Application of this model to thermal cycles is illustrated using the concept of the "kinetic strength" of the cycle. The techniques described may be generalised to a wide range of thermal processing.

1601 Process Modelling Applied to Age Hardening Aluminium Alloys

The age hardening aluminium alloys have been widely studied, to the exent that the underlying physical processes are well established. Kinetic models of precipitation, coarsening and dissolution are available, as are relations describing dislocation interactions with the precipitates. Recently attempts have been made to assemble complete process models for industrial treatments involving age hardening [1-4]. By this we mean a mathematical description of the relation between the process variables and the consequent properties, with a sound physical basis. Process models are relatively rare in the processing of solids - in contrast to many years of application in chemical engineering. The benefits are clear, however, - faster process development, optimisation of process and properties, and the potential for real-time control. This review paper brings together two process models, developed independently, which describe the microstructure and property evolution of age hardening aluminium alloys. Both projects initiated from an interest in predicting the strength loss in the heat-affected zones of welds in 6000 series alloys. However, they share a common philosophy which has wide applicability in thermal processing of aluminium, and other alloys (for example, in extrusion, hot forming, welding, laser processing, and heat treatment generally).

1601.01 General Methodology

The process model draws together established knowledge of the kinetics of microstructure evolution with dislocation behaviour, in order to determine the mechanical properties. For some thermal processing, appropriate heat flow calculations may also be required to predict the temperature-time history. An important feature of the method is to maintain a consistent level of accuracy in all of the components of the model. It is also desirable to keep the models as simple as possible, to make them quick and easy to implement, and accessible to all potential industrial users. These two considerations therefore suggest the use of approximate, analytical solutions within the scope of personal computers. A second general feature is the use of internal state variables to describe the microstructure. The breadth of application of this approach in materials science and engineering has been discussed recently by Ashby [5]. The evolution of the microstructure is captured mathematically in terms of differential variation of the state variables with time, for each of the relevant mechanisms. These equations are then integrated through the thermal history and coupled to the subsequent mechanical properties. Finally, as models of this type inevitably contain poorly-known constants, a means for calibrating the models to experimental data is required. For ease of use the aim is to base calibration of the models entirely on easily measured macroscopic quantities (such as strength) rather than requiring direct measurement of the state variables themselves, which is invariably laborious and expensive.

At the simplest level, the microstructure of an age hardening aluminium alloy is described by two state variables: the volume fraction of precipitate, f, and the number of particles per unit volume, N. The mean particle radius, r, may be used in preference to one of these as only two of the variables f, N and r are independent. In the early stages of isothermal ageing, or throughout a thermal cycle, both volume fraction and number of particles per unit volume (or mean radius) may evolve with time. In general therefore, microstructure evolution must be described in differential terms as follows, (taking f and N as the independent variables):

$$\frac{\delta f}{\delta t} = g_1(f, N, T...)$$

$$\frac{\delta N}{\delta t} = g_2(f, N, T...)$$
(1b)

A "response equation" is also required which describes the precipitation strength increment, $\Delta \sigma_n$, for the current values of f and N:

$$\Delta \sigma_p = g_3(f, N) \tag{1c}$$

Solution of these coupled equations generally requires stepwise integration in time. In the first instance, however, we assume that the volume fraction is constant and equal to its equilibrium value at the ageing temperature. Hence equation (1a) is equal to zero and the problem reduces to one with evolution of only one state variable, N(or r). The second model, presented later, takes a second simple situation - that of dissolution or precipitation of a constant number of particles, in which case the second differential equation is equal to zero and f becomes the single evolving variable. The modelling is introduced in this form to indicate the general state variable approach of which these models are special cases.

1601.02 Model for Isothermal Age Hardening

- Components of the model
- Calibration of the model
- Application of the model

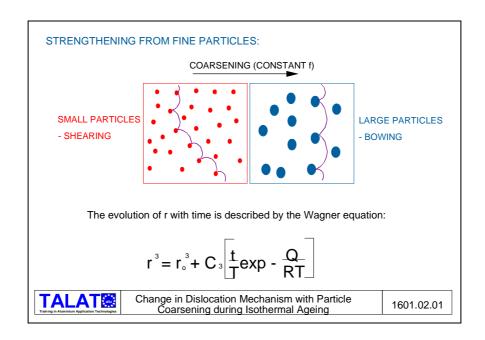
Components of the Model

A complete model for isothermal age hardening must include the following components:

- (a) the initial growth of a volume fraction of precipitate, and consequent changes in solute concentration;
- (b) the temperature dependence of the equilibrium volume fraction and, through this, the equilibrium residual solute concentration in the matrix;
- (c) precipitate coarsening;
- (d) the contribution of solute, shearable precipitates and non-shearable precipitates to the yield strength.

Simplified equations describing these components have been taken from established sources and are described in full in ref. [1]. Here we present only the essentials of the model - the development of the precipitation strength.

This is illustrated in **Figure 1601.02.01**: coarsening of a fixed volume fraction of precipitate leads to a transition from shearing of small, closely-spaced particles to bypassing of large, stronger particles.



Coarsening is described by the growth of the mean particle radius r with time. No attempt has been made at present to include changes in particle shape or in the distribution of particle size during coarsening, though it is acknowledged that these can be important effects. It is also assumed that a single precipitate type dominates mechanical behaviour throughout the whole ageing curve.

The contribution to the strength due to a volume fraction f of precipitates of mean radius r depends on the type of dislocation interaction. For shearing of precipitates, it is described by:

$$\Delta \boldsymbol{\sigma}_{ns} = c_1 f^{\frac{1}{2}} r^{\frac{1}{2}}$$

(2a)

while for particles which are bypassed it is:

$$\Delta \sigma_{pb} = c_2 \frac{f^{1/2}}{r}$$

(2b)

where c_1,c_2 (and subsequently c_3 and c_4) are constants for a given alloy system. These constants are combined and re-expressed later in terms of measurable quantities. Initially we consider isothermal ageing assuming the equilibrium volume fraction of precipitate for the ageing temperature exists throughout. Coarsening at fixed volume fraction may be described by the standard theory verified by numerous studies [6-10]. In this simple situation r is the only variable, and so there is no need for a differential expression. In integrated form, the evolution of r with time t is described by:

$$r^{3} = r_{0}^{3} + c_{3} \left[\frac{t}{T} e x p - \frac{Q}{RT} \right]$$
 (3)

where r_0 is the mean radius at t=0, T is the temperature and Q is an activation energy for ageing. The term in square brackets we call the "kinetic strength", P, of the thermal process. The more general use of the concept of the kinetic strength to characterise thermal cycles is described later. Examination of ageing curve data reveals that the value of P required to reach the peak over a wide range of temperature is often roughly constant. Hence we use this value, P_p , to normalise the kinetic strength (noting that $P/P_p=1$ at the peak). Over most of the ageing curve, the initial radius $r_0<<$ r, hence we may re-write equation (3):

$$r = c_4 \left(P/P_p \right)^{1/3} \tag{4}$$

Substituting for r in equation (2) leads to the expressions:

$$\Delta \sigma_{ps} = 2S_0 \left(P/P_p \right)^{\frac{1}{6}} \text{ and } \Delta \sigma_{pb} = 2S_0 \left(P/P_p \right)^{-\frac{1}{3}}$$
 (5)

Here, all the unknown constants are combined into a single parameter, S_0 , with dimensions of strength. Note in particular that $S_0^2 \alpha f$ - a feature which is exploited to find the temperature dependence of f. The net precipitation strength due to these

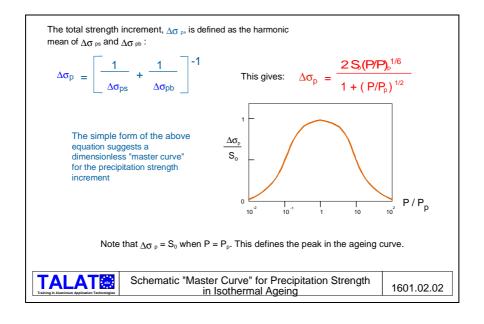
competing mechanisms will be dominated by the lower of the two. There is a smooth transition between shearing and bypassing due to the particle size distribution (a further microstructural variable which is ignored in the present model). A convenient mathematical form which gives the required behaviour is a harmonic mean, though others could be investigated:

$$\Delta \sigma_p = \left[\frac{1}{\Delta \sigma_{ps}} + \frac{1}{\Delta \sigma_{pb}} \right]^{-1}$$
 (6a)

Substituting from equation (5):

$$\Delta \sigma_{p} = \frac{2S_{0} \left(P/P_{p} \right)^{\frac{1}{6}}}{1 + \left(P/P_{p} \right)^{\frac{1}{2}}}$$
 (6b)

The simple form of equation (6b) suggests the existence of a dimensionless "master curve" for the precipitation strength; which is independent of temperature - this is illustrated schematically in **Figure 1601.02.02**. Note that the peak precipitation strength is equal to S_0 . The simple form of equation (6b) suggests the existence of a dimensionless "master



Calibration of the Model

The temperature dependence of the equilibrium volume fraction is contained in the parameter S_0 . Consideration of a solvus boundary on the phase diagram [1] leads to an expression for the temperature dependence of S_0 :

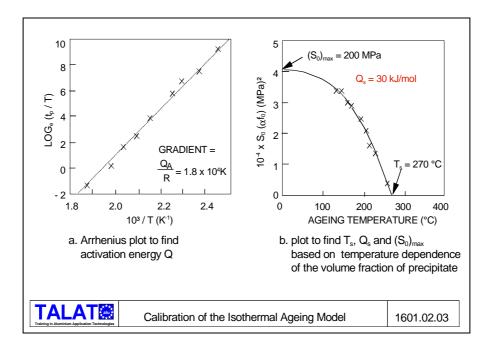
$$(S_0)^2 = (S_0)_{\text{max}}^2 \left[1 - \exp(Q_s/R) \left(\frac{1}{T} - \frac{1}{T_s} \right) \right]$$
 (7)

where $(S_o)_{\max}$ is the maximum value obtainable if all the solute precipitated out, Q_s is an enthalpy of solution, and T_s is the solvus temperature for the dominant precipitate formed during ageing. Values are therefore sought for five unknowns: P_p , Q, $(S_o)_{\max}$, Q_s and T_s .

The construction of the model has been done in such a way as to decouple the time variations due to coarsening, governed by Q and P_p , from the variations which depend only on the ageing temperature, that is, the equilibrium volume fraction described by S_0 .

Values for the unknown parameters are all found by calibrating the data to the peaks of the ageing curve. By finding the time to peak, t_p , at various temperatures P_p is evaluated directly. Noting that $P_p = const. = \left(t_p/T\right) \exp\left(-Q/RT\right)$, an Arrhenius plot of $\log_e(t_p/T)$ against 1/T gives a value for Q from the slope Q/R - an example is shown in **Figure 1601.02.03a** for alloy 6061 (data from ref. [11]). The measured peak strength includes contributions from other sources than precipitation (such as solute strengthening). By subtracting an appropriate allowance for other contributions to the peak strength, data for $\Delta\sigma_p$ (and thus S_0) may be found for various temperatures.

Figure 1601.02.03b shows a plot of $(S_0)^2$ against temperature, with equation (7) superposed using values for $(S_0)_{\max}$, Q_s and T_s which yield the best fit. Hence values for all five unknowns may be found from a subset of the data, after which the entire ageing curve at any temperature may be predicted.

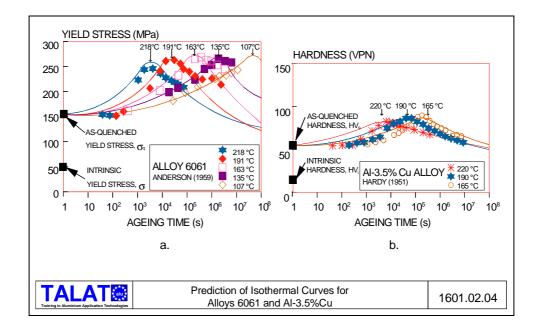


The formulation of the model has a number of general features which are worthy of note:

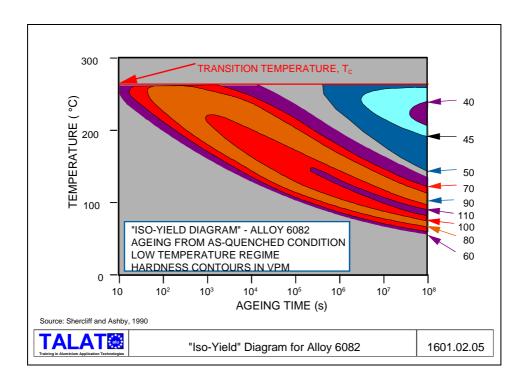
- (a) the model has been constructed such that, where possible, the unknown constants are combined into parameters which have physical meaning. Hence, when values for these parameters are found by calibration to experimental data, we have a check that the values are reasonable:
- (b) construction of the equations in this way is greatly facilitated by the use of dimensionless variables, such as P/P_p ; in particular, raising a dimensionless variable to a power means that the premultiplying constant does not depend on the value of the power.

Application of the Model

The model described above, with some refinements not detailed here, has been applied to alloys 6061 and 6082, and to five binary aluminium-copper alloys (full details in refs.[1,2]). **Figures 1601.02.04a** and **b,** shows the model compared with the ageing curves for two of these alloys.



The agreement is good around the peak and in some cases beyond. It is clear, however, that the approximate treatment of the initial precipitation proposed in ref.[1] and used to construct the curves could be improved - the data show a more marked "incubation period" before rising to peak strength. The essentials of isothermal ageing have, however, been captured to a useful extent by the model. The model may now be used as a tool for predicting the effect of treatments for which data were not provided, or in devising novel "process diagrams" for isothermal ageing. **Figure 1601.02.05** shows an example - an "iso-yield diagram" which shows hardness contours on a plot of temperature and ageing time.



1601.03 Models for Isothermal Reversion and Precipitation

- Reversion model
- Precipitation model
- Natural ageing model

The second set of models were developed independently, using very similar techniques, with the same goal of simplicity at a useful level of accuracy. These models addressed directly the softening reactions taking place in the heat-affected zone of welds in 6082-T6 alloys. In this material the heating cycle causes the hardening β " particles to undergo reversion, then on cooling some of the solute may precipitate out as essentially non-hardening β ' particles. In the context of welding, the importance of this precipitation during cooling is the effective loss of solute for subsequent natural ageing, and thereby strength recovery. The models are not limited to welding, however, but may be applied to any process involving reversion of T6 material, and precipitation on cooling a solid solution. Full details of the models, and their application to welding are given in refs. [3,4]. Here, we summarise the main features of the models, and discuss their application to thermal cycles in section 1601.04.

Reversion Model

This model assumes a close-packed 2-D array of parallel rod-shaped particles, each dissolving in a cell of diameter equal to the particle spacing. Hence the number of particles per unit volume is constant, and the volume fraction and particle radius do not vary independently (as $f\alpha r^2$). The problem is therefore described by a single state variable, f. It is assumed that the precipitation strength increment, $\Delta\sigma_n$, is given by:

$$\Delta \sigma_p = \left(\Delta \sigma_p\right)_{\text{max}} \left(f/f_0\right)^m \tag{8}$$

where m is assumed equal to unity. The previous equation for strength due to particle shearing (equation (2a)) with $f \alpha r^2$ gives the value of m=3/4, which is a minor discrepancy. Note also that calibration to a measured maximum strength increment has eliminated an unknown proportionality constant. Simple diffusion theory shows that the volume fraction falls from its initial value, f_0 , according to the equation:

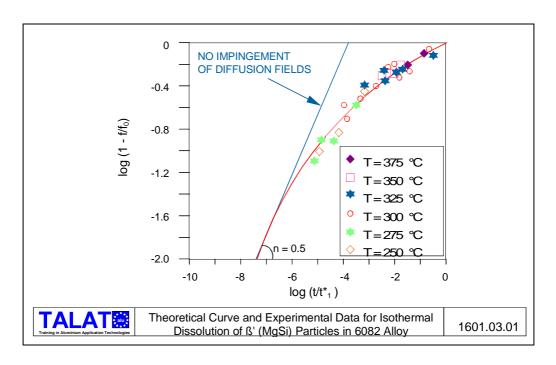
$$f/f_0 \approx 1 - \left(t/t_1^*\right)^n \tag{9}$$

where t_1^* is the time taken for complete dissolution at the temperature concerned. The variation of t_1^* with temperature is given by

$$t_1^* = t_{r1}^* \exp\left[\left[\frac{Q_s}{nR} + \frac{Q_d}{R}\right]\left[\frac{1}{T} - \frac{1}{T_{r1}}\right]\right]$$
 (10)

In common with the first model, this model also exploits some good modelling techniques, noted in section 1601.02 "Calibration of the Model". For example: (i) the use of a dimensionless time eliminates an unknown kinetic constant which premultiplies t and t_1^* in the derivation of equation (9); (ii) raising the dimensionless time to a power n means that the premultiplying constant, here unity, is independent of the value of n and is itself also dimensionless; (iii) the form of equation (10) eliminates further unknown kinetic constants and may readily be calibrated using an experimental time t_{r1}^* at a reference temperature T_{r1} .

Figure 1601.03.01 shows the variation in f/f_0 with time (on log axes) from a range of isothermal experiments, using hardness measurement to evaluate $\Delta \sigma_p$ and $\left(\Delta \sigma_p\right)_{\rm max}$, and evaluating f/f_0 using equation (8). The curve (equation (9)) extrapolates back to a slope of 0.5 (the exponent n) for the case of the early stages of dissolution before impingement of the diffusion fields. The exponent n is seen to fall to lower values when the proportion dissolved is higher.



Precipitation Model

On cooling a solid solution of 6082 from high temperature, or by quenching and holding isothermally, precipitation of β ' particles will occur. The fraction of solute precipitated, X_e , may be described by an Avrami expression:

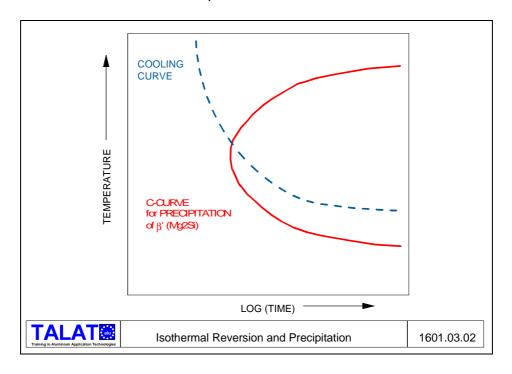
$$X_{\rm e} = 1 - \exp\left(-t/\tau\right) \tag{11}$$

where τ is a temperature-dependent time constant, and taking an exponent on t/τ equal to unity. The temperature-dependence of τ is found by considering the kinetics of the C-curve for β ' precipitation, as in **Figure 1601.03.02**. The transformation rate depends on the product of a driving force, and a thermal activation. Long transformation times occur at low undercoolings due to the weak driving force, while at low temperatures the thermal activation is small. The shortest transformation times thus occur at an intermediate temperature.

The β ' particles are essentially non-hardening, so we are most interested in the remnant solute after a given treatment time, X_{ss} , equal to $1-X_e$. Manipulation of equation (11), coupled to a description of the shape of the C-curve [3], yields an equation of the form:

$$X_{ss}^{2} = (0.95)^{(t/t_{2}^{*})}$$
 (12)

The value 0.95 stems from defining the start of precipitation (as described by the C-curve) as occurring at a value of $X_e = 2.5\%$. As before, a dimensionless time eliminates unknown parameters, and t_2^* is defined as a function $F(t_{r2}^*, T_{r2}, T, \text{kinetic constants})$ from the kinetics of the C-curve for β '.



Natural Ageing Model

In the welding problem, it is of most interest to predict the final strength of the HAZ, after any recovery of strength has occurred by natural ageing. The time evolution of strength during natural ageing has not therefore been modelled, but simply the maximum value which can be obtained if the solute retained at the end of the weld cycle is left to age. A maximum precipitation strength increment by natural ageing was measured experimentally, $\left(\Delta\sigma_p\right)_{\max,na}$, obtained by precipitation from 100% solid solution (that is, $X_{ss}=1$). The strength increment after natural ageing, following on from precipitation of β ' on cooling, which left a fraction X_{ss} in solution is given by:

$$\Delta \sigma_p = \left(\Delta \sigma_p\right)_{\text{max } nq} X_{ss}^2 \tag{13}$$

This follows from the volume fraction of precipitate formed on natural ageing being proportional to X_{ss}^2 , and the strength increment being proportional to f (equation (8)). Myhr and Grong [3] successfully validated the coupled models for β 'precipitation followed by natural ageing using measurements of both hardness and electrical resistivity.

1601.04 Application to Thermal Cycles

The set of models described in section 1601.03 were applied to weld thermal cycles, in which the temperature varied continuously with time. The models are simple in the sense that there is only one state variable, whose isothermal evolution in time has been modelled for the processes of reversion and subsequent precipitation. For a thermal cycle we must integrate an evolution equation, df/dt. This is, therefore, a simple case of the general formulation described in section 1601.01. We now invoke the related concepts of an isokinetic reaction and the kinetic strength of a thermal cycle.

A reaction is said to be isokinetic if the increments of transformation in infinitesimal isothermal time steps are additive. Christian [12] defines this mathematically by stating that a reaction is isokinetic if the evolution equation for some state variable, X, may be written in the form:

$$\frac{dX}{dt} = \frac{G(X)}{H(T)} \tag{14}$$

For a given thermal history T(t) this essentially means that the differential equation contains separable variables X and t. This criterion may be applied to the models derived above. Considering reversion first, we may re-write equation (9) as:

$$I_1 = (1 - f/f_0)^{1/n} = t/t_1^* \tag{15}$$

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Differentiating:

$$\frac{df}{dt} = \frac{df}{dI_1} \left(1/t_1^* \right) \tag{16}$$

 df/dI_1 depends only on f, while t_1^* depends only on temperature so the additivity requirement, equation (14), is satisfied. So when the temperature varies with time, we replace t/t_1^* in equation (9) by dt/t_1^* and integrate over the thermal cycle, giving:

$$I_{1} = \int_{0}^{t} \frac{dt}{\mathcal{R}} = \left(1 - f/f_{0}\right)^{\frac{1}{2}} \tag{17}$$

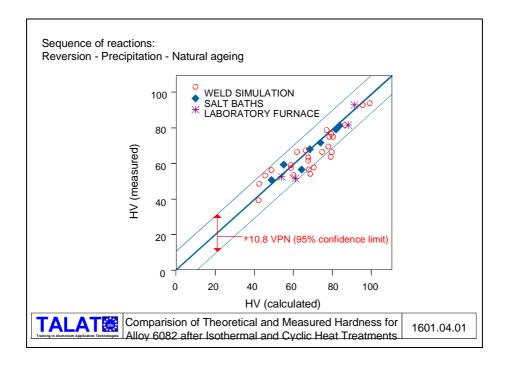
This integral is called the kinetic strength of the thermal cycle, with respect to reversion. The precipitation strength following a heating cycle which causes reversion is found by:

- (a) evaluating I_1 numerically;
- (b) replacing t/t_1^* with I_1 in equation (9), yielding a value for f/f_0 from the curve of **Figure 1601.03.01**;
- (c) substituting for f/f_0 in equation (8).

A similar argument may be followed to show that the evolution of the remnant solute fraction, X_{ss} , in the precipitation model is also isokinetic, so a second kinetic strength with respect to β ' precipitation, I_2 , may be found. The value of this integral for the cooling cycle is substituted for t/t_2^* in equation (12) to yield the value of X_{ss} used in the subsequent natural ageing calculation. Myhr and Grong applied their combined models to a wide range of heat treatment cycles - the measured and predicted strength (in this case hardness) are compared in **Figure 1601.04.01**. The prediction of the models in almost all cases lies within a 95 % confidence limit. Similar good agreement of the model and data is shown in **Figure 1601.04.02**, which shows the hardness profile across a weld in 6082-T6 plate.

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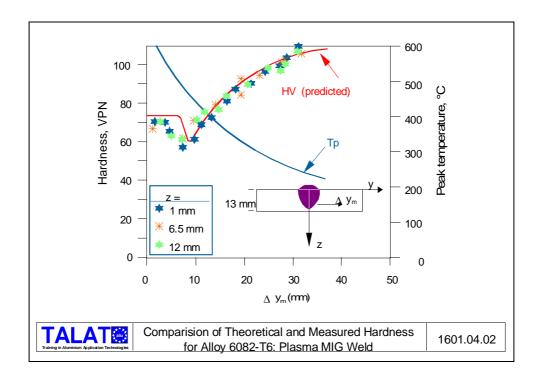
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Note also that the kinetic strength with respect to coarsening, P, defined for isothermal ageing in section 1601.01 is essentially the same, but in dimensional form:

$$P = \int_{0}^{t} \frac{1}{T} \exp\left(\frac{Q}{RT}\right) dt = \left(\frac{t}{T}\right) \exp\left(\frac{Q}{RT}\right)$$
 (18)

The integral is trivial as T is not a function of time. It is important to realise that modelling of coarsening using a kinetic strength integral evaluated over a thermal cycle is potentially in error, as the volume fraction is not then constant - that is, we have evolution of two state variables. Provided the volume fraction varies little during the cycle, the kinetic strength may be used as a first estimate of the effect of the cycle. This method was used by Shercliff and Ashby [2], who assumed that the volume fraction was constant and equal to the value for the peak temperature of the cycle. As a final note it should be borne in mind that the isokinetic approximation may break down due to the influence of other mechanisms - for example, the retention of a non-equilibrium concentration of vacancies on quenching, or local enhancement of diffusion due to high dislocation densities around inclusions.



1601.05 Conclusions and Future Work

- 1) Process modelling methods have been applied successfully to some common kinetic processes in age hardening aluminium alloys. Considerable progress can be made using simple analytical solutions. The potential benefits of such models in terms of process optimisation and control, and alloy development are clear.
- 2) Judicious construction of the equations makes full use of both dimensionless parameters, particularly in power law relationships, and calibration techniques which eliminate the poorly known kinetic constants. Calibration of the model is achieved using readily observable macroscopic quantities which reflect the state variables, rather than direct observation of the variable quantities themselves, which is inherently laborious, involving microscopy or chemical analysis. It is however acknowledged that correct identification of mechanisms at the outset, and subsequent testing of the models can benefit greatly from application of high resolution microscopy.
- 3) The general state variable method is a useful starting point in constructing models of this type, particularly when thermal cycles or continuous cooling are of interest. Complex treatments involving evolution of more than one state variable will generally require stepwise solution of a coupled set of differential equations. The models illustrated here are based on evolution of single state variables. All of the isothermal models may thus be formulated directly in integrated form. In addition the reversion and precipitation models, applied to a thermal cycle, may be integrated in a single step, using the kinetic strength of the cycle. This simplification is only possible in cases for which the evolution equation is isokinetic.

The modelling methods described, and the specific models themselves, may be readily applied in a wide range of treatments in the aluminium industry. Work is in progress to model the quench sensitivity of 6000 series extrusions, using a combination of the models for precipitation on cooling, and isothermal artificial ageing. This will lead to a more general application of the theory of C-curves for continuous cooling in heat treatment of aluminium alloys. Refinement of the predictions of strength loss in a weld HAZ remains of some interest, given the present empirical nature of design codes for aluminium weldments. Initial investigations of the applicability of the models to 7000 series material have identified the greater complexity of this series compared to the 6000 series, but further work on 7000 series alloys is intended. The methods may also be successful in modelling recovery and recrystallisation during thermal processing of 5000 series alloys.

1601.06 Literature

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