EFFECT OF SOLUTE DRAG ON SOFTENING DURING ANNEALING OF COMMERCIAL PURITY ALUMINIUM; EXPERIMENTAL INVESTIGATIONS AND MODELLING TREATMENT

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ABSTRACT A model for softening during annealing of rolled Al-sheets is presented. The model includes recovery (subgrain growth and annihilation of dislocations) and recrystallisation as softening mechanisms. Model predictions have been compared with experimental results for cold rolled and annealed commercial purity aluminium with various amounts of Fe and Si and also additional Mn. Isothermal annealing in a salt bath furnace was undertaken at temperatures in the range 200-400°C and the strength was measured as a function of annealing time by tensile testing. The model predictions were in good agreement with experimental results. The predictions confirm that thermal activation of solute atoms is rate-controlling for recovery, and variations in solid solution levels can be handled by the model. The results demonstrated that annihilation of dislocations within subgrain interiors gives a significant contribution to the softening, and must be included in the model in order to give satisfactory predictions.

Keywords: *Rolling, recovery, recrystallisation, modelling*

1. INTRODUCTION

Rolled aluminium sheets are usually annealed in order to obtain desired mechanical properties like strength and formability. During the annealing cycle the material softens, while the ductility increases. Although the loss of strength is usually undesirable, the annealing sequence must be performed in order to fulfil the requirements for subsequent forming operations. The purpose of the present work has been to develop a model for predictions of mechanical strength of rolled and annealed Al-sheets. The model concept is based on previous modelling approaches, but has been refined and further developed. A softening model will be a useful and industrially relevant tool for determination of annealing conditions (annealing times and temperatures) in order to obtain requested mechanical properties for a specific product application, for decision of alloy chemistry for products with special requirements to softening behaviour and for handling products which have been subjected to deviations in alloy chemistry or processing during production.

2. MODEL CONCEPT

Several work hardening models [1-4] have been developed over the last years where the strength of a deformed material is expressed through the substructure parameters δ (subgrain size) and ρ (the dislocation density in the subgrain interiors) in the following way:

$$R_{p0.2} = R_{FLP} + \alpha_1 MGb\sqrt{\rho} + \alpha_2 MGb\frac{1}{\delta} \qquad (1)$$

where G is the shear modulus, b is the burgers vector, M is the Taylor factor, α_1 (of the order 0.3) and α_2 (of the order 2.5) are constants and R_{FLP} is the friction stress due to precipitates and elements in solid solution.

During annealing the subgrains will grow and internal dislocations will annihilate. These recovery reactions lead to a reduced strength of the material. In addition, recrystallisation may occur, which also contributes strongly to the softening. The simplest way of generalising Eq. (1) to a situation during annealing, will be as follows:

$$R_{p0.2}(T,t) = R_{FLP} + \left[\alpha_1 MGb\sqrt{\rho(T,t)} + \alpha_2 MGb\frac{1}{\delta(T,t)}\right][1 - X(T,t)] \qquad (2)$$

where X is the fraction recrystallised. The friction stress (R_{FLP}) will correspond to the flow stress in a fully recrystallised condition of the material. To a first order approximation the friction stress is assumed to be independent of time and temperature. The factor (1-X) is simply an expression which linearly scales the strength with the fraction unrecrystallised in the material. At a fraction X=0 the strength corresponds to the flow stress in the as-rolled condition and for X=1 (fully recrystallised) the flow stress corresponds to the friction stress.

A softening model for heavily deformed aluminium alloys now needs relationships for annihilation of dislocations within the interior of the subgrains, subgrain growth and recrystallisation. In commercial purity aluminium there will always be a small fraction of Fe (and possibly other elements) in solid solution, which may have a considerable effect on the softening process. In a case where the recovery processes are assumed to be determined by thermal activation of solute atoms, the evolution of internal dislocation density, ρ, and subgrain size, δ, will be given as follows [5,6]:

$$\frac{d\rho}{dt} = -\nu_D b B_\rho \rho^{3/2} \exp\left(-\frac{U_g}{kT}\right) 2\sinh\left(\frac{A_\rho G b^4 \sqrt{\rho}}{kT}\right), \qquad A_\rho = \varpi_\rho c_{ss}^{-2/3} \qquad (3)$$

and

$$\frac{d\delta}{dt} = \nu_D b B_\delta \rho^{3/2} \exp\left(-\frac{U_g}{kT}\right) 2\sinh\left(\frac{A_\delta G b^4}{\delta kT}\right), \qquad A_\delta = \varpi_\delta c_{ss}^{-2/3} \qquad (4)$$

where U_g is the activation energy for recovery (i.e. activation energy for diffusion of solute elements), ν_D is the Debye frequency, b is the Burgers vector, B and ϖ are alloy-dependent constants, k is Boltzmann's constant and c_{ss} is the at% of elements in solid solution. Information about the strength in the as-rolled condition (i.e. X=0 in Eq. 2) and the fully recrystallised condition (X=1) is very useful to determine constants in the model. If this information is available for the alloy of interest (measured or predicted by other models) in addition to information about the respective contributions from internal dislocations and subgrains to the as-deformed strength (expressed through a factor f_ρ that is the strengthening contributions from internal dislocations), the parameters ρ_0 (initial dislocation density), δ_0 (initial subgrain size) and R_{FLP} are easily determined.

The recrystallisation kinetics are calculated by a standard Avrami equation of the following form:

$$X(T,t) = 1 - \exp\left[-(t/\tau_r)^n\right], \qquad \tau_r = \tau_r^0 \exp(U_r/RT) \qquad (7)$$

where U_r is the activation energy for recrystallisation, τ_r^0 is the pre-exponential of the relaxation time for recrystallisation, τ_r, and n is the Avrami exponent.

3. MODELLING CASES

Softening curves of several commercial purity aluminium alloys were modelled. Their chemical compositions and amounts of elements in solid solution are given in Table 1. The values for the solute content were determined by TEP (Thermo-Electric Power) or electrical conductivity measurements in combination with phase diagram calculations, but can only be considered as best estimates. Notice that alloy 1145 was processed in such a way as to give 2 different levels of Fe in solid solution. All alloys were cold rolled to a strain of 3 and tensile specimens were machined from the rolled sheets. The samples were isothermally annealed in salt bath for different times at various temperatures and tensile testing was undertaken in order to determine the softening curves.

Table 1: Chemical compositions and amounts of elements in solid solution in the modelled alloys. The given amounts in solid solution are either determined by TEP (Thermo-Electric Power) or electrical conductivity measurements in combination with phase diagram calculations.

Alloy	wt% Si	wt% Fe	wt% Mn	C_{ss} [wt%]
1070	0.05	0.17		0.005 (Fe)
1070+Mn	0.07	0.14	0.89	0.2 (Mn)
1050	0.11	0.25	0.01	0.006 (Fe)
1145A	0.09	0.43		0.03 (Fe)
1145B	0.09	0.43		0.006 (Fe)

There are several material parameters in the model that must be determined: (i) parameters relating substructure and strength, (ii) parameters related to recovery and (iii) parameters related to recrystallisation. Material strength in the as-rolled and fully recrystallised conditions were used as input, and assumptions must be made in each case for the relative contributions to the mechanical strength from internal dislocations and subgrains in the as-rolled condition (the contribution from internal dislocations will typically be of the order 20% for commercial purity aluminium, i.e. f_ρ=0.2). Based on this assumption and knowledge about the as-rolled and fully recrystallised strength all basic parameters in Eq. (2) and initial values of subgrain size and dislocation density can be determined. The other modelling parameters were determined by tuning to the experimental data (their values will be given below).

4. MODEL PREDICTIONS AND COMPARISON WITH EXPERIMENTAL RESULTS

Alloy 1050; effect of internal dislocations:

It is usually assumed that most of the material strength is due to dislocations stored in subgrain boundaries, while the effect of the internal dislocations is often ignored. The 1050 alloy was used to investigate the effect of internal dislocations on the softening curves. A comparison between model predictions and experimental data is shown in Fig. 1. In Fig. 1a both annihilation of internal dislocations (Eq. 3) and subgrain growth (Eq. 4) have been included, while internal dislocations have been ignored in Fig. 1b (i.e. f_ρ=0). The following model parameters were found to give the best agreement with experimental data: U_g=194kJ/mol, B_ρ=1.5x10^8, ϖ_ρ=2.3, B_δ=7x10^1, ϖ_δ=0.05, f_ρ=0.2, ρ_0=1.74x10^{13}m^{-1}, δ_0=0.5μm, U_r=194kJ/mol, n=2.2, τ_r^0=5x10^{-15}. The activation energy of 194kJ/mol corresponds with that of diffusion of Fe [7], i.e. the thermal activation of solute Fe atoms is rate controlling for the recovery processes. As can be seen from the figure, the model which includes both annihilation of dislocations and subgrain growth gives a very good agreement with experimental results. The model which only considers subgrain growth gives

significantly poorer model predictions in the recovery part of the softening curve. If the constants in the poor subgrain growth model are chosen so as to predict the initial softening correctly at low temperatures, these parameters give a too fast subgrain growth at higher temperatures and longer annealing times. On the other hand, if the subgrain growth model is fitted at high temperatures and long annealing times, the model fails initially for lower temperatures. This demonstrates that annihilation of internal dislocations contributes significantly to the softening in the initial part of the recovery period. The good agreement confirms that the model concept is very reasonable.

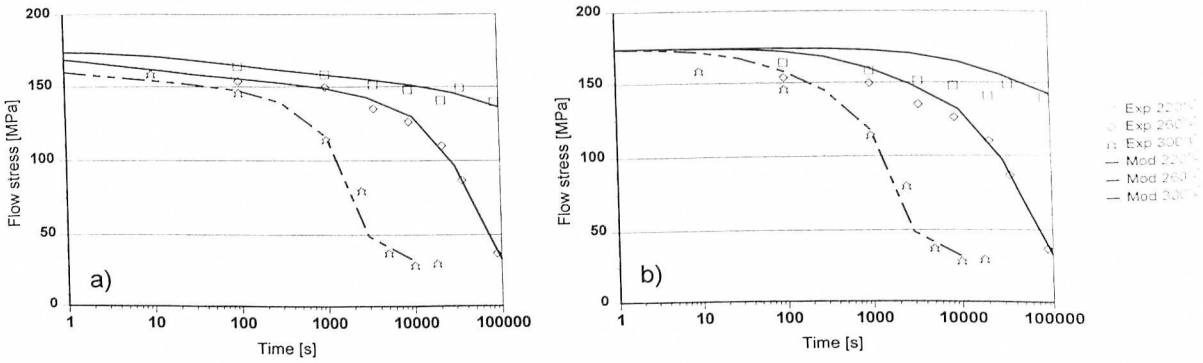

Fig. 1: Model predictions compared with experimental data of alloy 1050: a) model including subgrain growth and annihilation of internal dislocations, b) model including only subgrain growth.

Alloy 1145; effect of Fe in solid solution:

The two different amounts of Fe in solid solution in alloy 1145 enables a test of the model's ability to handle differences in solute concentration. It would be expected that a higher level of Fe would retard the kinetics of recovery, as is also predicted by the model and confirmed by the experimental data, see Fig. 2. Figure 2a and 2b show the cases of low and high Fe-levels in solid solution, respectively. Good agreement between experimental data and model predictions was found. For both alloy variants the following model constants were used: U_g=194kJ/mol, B_ρ=1.5x10^8, ϖ_ρ=2.3, B_δ=3x10^4, ϖ_δ=0.05, f_ρ=0.25, ρ_0=1.5x10^{13}m^{-1}, δ_0=0.72μm, U_r=194kJ/mol, n=2.2, τ_r^0=1x10^{-15} (both annihilation of dislocations and subgrain growth were considered).

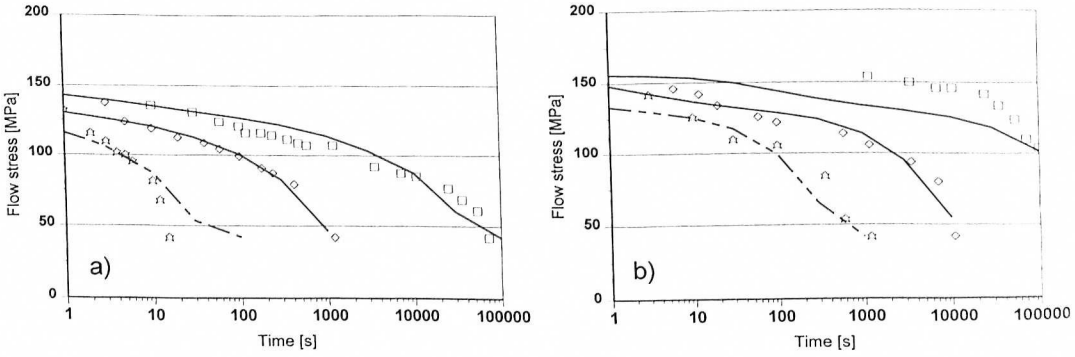

Fig. 2: Model predictions compared with experimental data of alloy 1145: a) alloy variant with 0.006wt% Fe in solution and b) alloy variant with 0.03wt% Fe in solid solution (for details on experimental data, see Ref. 8).

However, the relaxation time for recrystallisation had to be slightly increased (τ_r^0=2x10^{-14}) for the material with the highest amount of Fe in solid solution, as the recrystallisation kinetics were much slower for this alloy. This is believed to be due to precipitation of small Fe-particles during annealing, which slow down the nucleation and growth of recrystallised grains. Precipitation was confirmed by measurements of electrical conductivity [8]. As a retarding Zener drag from precipitates is not included in the model, there will be some discrepancy for the recrystallisation part of the softening curve. The higher solute content also gave a slightly different as-rolled substructure: ρ_0=1.75x10^{13}m^{-1} and δ_0=0.66μm. Again, an activation energy corresponding to thermal activation of solute Fe was found. The model parameters were slightly different from the 1050 case above. This difference is no surprise, as the data on alloy 1145 have been recalculated from hardness measurements (using the conversion $R_{p0.2}$=4VHN-46, where VHN is the Vicker's hardness and the flow stress is in units of MPa) and a quite different material supplier, pro-processing procedure and laboratory facilities had been used. However, identical parameters could be used for the two 1145 variants (except for τ_r^0). Their different behaviour is handled by the model through the difference in solute concentration (c_{ss} was about 5 times higher in the most soluble variant).

Alloy 1070; effect of Mn addition:

Figure 3a and 3b show model predictions and experimental data with and without addition of Mn, respectively. The standard 1070 alloy (Fig. 3a) was modelled with the following parameters: U_g=194kJ/mol, B_ρ=1x10^{11}, ϖ_ρ=2.3, B_δ=7.5x10^3, ϖ_δ=0.05, f_ρ=0.12, ρ_0=4.8x10^{12}m^{-1}, δ_0=0.51μm, U_r=194kJ/mol, n=2.2, τ_r^0=1x10^{-16}, which gave very good agreement with experimental results. The model constants are very comparable to those of the 1050 alloy above. An attempt was made to use the same parameters for the alloy with additional Mn, just by changing the solute content (and as-rolled flow stress) as model input. This attempt was not successful. This is no surprise, as activation of solute Mn-atoms will now be rate-controlling for the recovery processes. By changing the activation energy from 194kJ/mol to 240kJ/mol, the model predictions fitted the data well in the initial part of the softening curve. Still, the parameters for subgrain growth had to be slightly changed (B_ρ=2.6x10^{12}, ϖ_ρ=2.3, B_δ=1.95x10^5, ϖ_δ=0.22, f_ρ=0.25, ρ_0=2.63x10^{13}m^{-1}, δ_0=0.54μm). It is reason to believe that this is partly due to uncertainties in the calculation of the amount of elements in solid solution.

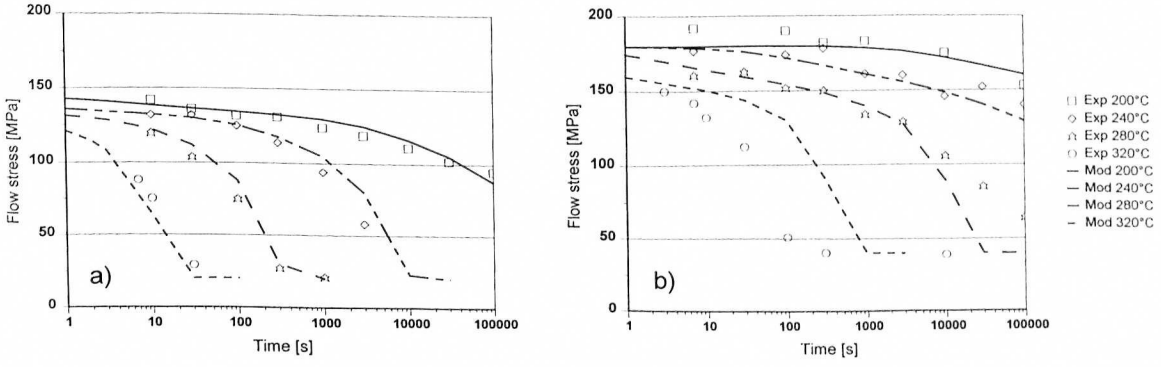

Fig. 3: Model predictions compared with experimental data (for details on experimental data, see Ref. 6): a) alloy 1070 (activation energy of 194kJ/mol) and b) 1070 with addition of 0.9wt% Mn (activation energy of 240kJ/mol).

The recrystallisation part of the softening curves are not so well described by the model for the Mn-containing alloy. As can be seen from the experimental results, the slopes of the softening curves are rather different for the temperatures of 280°C and 320°C, i.e. the Avrami-exponent is different, while the model assumes a constant Avrami-exponent of 2.2. This effect is most likely due to precipitation of Mn-particles during annealing, leading to a retarding Zener drag which is larger, the lower the temperature. This was confirmed by measurements of electrical conductivity [6]. It is even seen that the strength increases for the lowest annealing temperature, due to the formation of small Mn-clusters. As such effects are not incorporated in the model, it can not be expected that the model is able to predict such a behaviour. It will take some time before the precipitates form. This explains why the initial parts of the softening curve (annihilation of dislocations) is well described, but not the subgrain growth and recrystallisation regimes which are more strongly influenced by the precipitates. Notice also that the contribution from internal dislocations is higher in the Mn-containing alloy. The physical explanation of this is that the higher amount of solute atoms leads to a higher rate of internal storing of dislocations (dislocations are trapped by single solute atoms). In the model this was handled by changing the parameter f_p to 0.25.

5. CONCLUDING REMARKS

The attempts to model mechanical properties of rolled and annealed sheets have demonstrated that the present model concept for softening during annealing is reasonable. Thus, thermal activation of solute atoms appears to be rate-controlling for growth of internal dislocations in networks and subgrain growth. It was shown that annihilation of internal dislocations in subgrain interiors must be included, in addition to subgrain growth, in order to properly describe the recovery softening. The model is also able to predict differences between commercial purity aluminium alloys with various chemical compositions and solute levels, although a proper calculation or measurement of solute content is difficult and inaccurate. Several model parameters had to be changed when applying the model to different alloys and experimental data. The reason for this is to a large extent believed to be uncertainties in calculating the exact amount of elements in solid solution. Some of the softening curves were recalculated from hardness measurements, and some of the experimental data were taken from the literature, i.e. material suppliers, pro-processing procedures and laboratory facilities were quite different. Addition of Mn was handled by the model, but a concept for precipitates must be included for an improved description of softening in Al-Mn alloys. The model has so far mainly been applied to commercial purity aluminium, but will in near future be extended to other typical non-heat-treatable Al-alloys.

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