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A comparative study of the stress relaxation in aged and un-aged high-purity aluminium polycrystals

M. Z. BUTT, M. ZUBAIR, I. UL-HAQ
Metal Research Laboratories, Department of Physics, P.O. Box 1072, Government College, Lahore-54000, Pakistan

Stress relaxation in 99.996% aluminium polycrystals of average grain-diameter 0.30, 0.42 and 0.51 mm, annealed at 500°C and aged for six months at room temperature, has been studied as a function of initial stress level from which relaxation at constant strain was allowed to start. Data were also obtained with annealed but un-aged aluminium specimens of the same purity and grain size for comparison. The grain size has no notable effect on the strength parameters and stress-relaxation rate in both aged and un-aged aluminium. The room-temperature ageing causes significant increase in the yield stress, while tensile strength and fracture stress remain un-effected. The intrinsic height of the thermally-activable energy barrier (1.64 eV) evaluated for aged aluminium is comparable with that (1.94 eV) for un-aged aluminium, and is of the order of magnitude for recovery processes. In aged specimens, the relaxation rate at a given stress level is 30% larger and associated activation volume is accordingly smaller than that in un-aged specimens. This is most probably due to the diffusion of vacancies and/or residual gaseous and metallic impurity atoms to the cores of edge dislocations in aged specimens.

1. Introduction

It is well established [e.g. 1–12] that the mode of solute distribution in alloy crystals has a marked effect on their mechanical properties, such as internal friction, stress relaxation, work hardening, creep, temperature and concentration dependences of yield stress etc. For example, in stress-relaxation measurements made with polycrystalline copper and alpha-brasses containing 10 to 35 at.% Zn, Feltham [2] observed a peak at about 26 at.% Zn in the concentration dependence of the relaxation rate at a given temperature in the range 77 to 291 K. He ascribed it to the local ordering near jogs, resulting in a lowering of their misfit energy, and hence facilitates the migration of dislocations. Similarly, Demirskiy *et al.* [5] observed an unexpected appreciable increase in the concentration dependence of the critical resolved shear stress τ of Cu-Al alpha solid-solutions ($c = 1$ to 14 at.% Al) near the solubility limit. They attributed the deviation from parabolic τ/c curve in the case of fairly concentrated solid-solutions to the deviation from random distribution of solute atoms, which might well occur in specimens held at room temperature for rather long time.

Since point defects, like vacancies and residual impurities etc, are always present in nominally 'pure' metal crystals, it was considered by Butt and Khwaja [13] to be of interest to study any possible effect of room-temperature ageing on the mechanical response, e.g. yield, tensile and fracture stress etc, of 99.996% aluminium polycrystals. They found that the yield stress of Al specimens annealed at 500°C and then 'aged' for six months at room temperature was nearly 30% higher than that of annealed but un-aged specimens. This was ascribed to the diffusion of vacancies and/or residual impurity atoms to the cores of edge dislocations during ageing, which pinned the dislocations and made their movements rather difficult. There was however no notable effect of ageing on the tensile strength and fracture stress, probably due to the large strains involved.

Recently Butt *et al.* [14] observed that natural ageing of high-purity aluminium polycrystals (mean grain-diameter: 0.25, 0.36 and 0.47 mm) causes a significant increase (≈30%) in the strain-rate sensitivity $\Delta\sigma$ of a given flow stress σ at temperature T. Consequently the associated activation volume $v_\sigma = kT\,(\Delta \ln \dot{\varepsilon}/\Delta\sigma)_T$, where k is the Boltzmann constant and $\dot{\varepsilon}$ is the tensile strain rate, decreases by the same order in aged aluminium compared with that in un-aged one. Grain size has however no effect on $\Delta\sigma$ or v_σ in both the cases. The main object of the present study was to investigate the effect of ageing, if any, on the stress relaxation behaviour of high-parity aluminium polycrystals. Another objective of this work was to elucidate the rate process of stress relaxation in both un-aged and aged aluminium, and its dependence on grain size.

2. Materials and methods

Grade-1 polycrystalline aluminium obtained from Johnson, Matthey & Co. Ltd, London, was in the form of rods of 30 cm length and 5 mm diameter. The mean grain-diameter was 0.12 mm, and the main metallic impurities were: Mg (20 ppm), Cu (10 ppm), Fe (8 ppm), Si (3 ppm) and Ca (1 ppm). Specimens 7.5 cm long were cut from the as-received rods. They were divided into three batches, each being sealed in a pyrex tube evacuated to 1.3 mPa (10^{-5} torr), and were annealed at 500°C for 30, 60 and 90 min to obtain mean grain-diameter 0.30, 0.42 and 0.51 mm, respectively. Half of the specimens of each grain size were deformed at room temperature just after the heat-treatment, while the remaining ones were allowed to age at room temperature for six months prior to deformation.

Both aged and un-aged specimens were deformed in tension at a strain rate of $4.6 \times 10^{-3}s^{-1}$ (cross head speed 10 mm/min) in a Universal Materials Testing Machine (Model 1195 Instron Ltd. UK) at room temperature. A record of load versus time was obtained from a 100 kN load cell, and output of the load cell amplifier was calibrated prior to deformation of the specimens. During the deformation leading to fracture (max. tensile strain 35 to 55%), straining was frequently interrupted by arresting the cross head to observe stress relaxation at constant strain. The chart of the load-time recorder was driven at a speed of 100 mm/min, which speed enabled one to attain adequate resolution of stress changes with the passage of time. The full scale load range used was 2 kN. Stress-strain curve for un-annealed specimen of mean grain-diameter 0.12 mm (max. tensile strain 15%) was also obtained in tension. The full scale load ranges used were 2 kN and 5 kN in this case. This helped to study the effect of annealing on the strength of as-received aluminium as well.

3. Results and discussion

The data points in Fig. 1 denote the measured values of yield stress (circles), ultimate tensile stress (squares) and fracture stress (triangles) of both as-received ($D = 0.12$ mm) and annealed ($D = 0.30$, 0.42 and 0.51 mm) 99.996% Al polycrystals. In the case of annealed specimens, the filled symbols refer to the room-temperature aged specimens while empty symbols stand for un-aged specimens; absence of grain size effect in each category of stress parameters is evident. One can also readily note that the yield stress of un-aged specimens (≈13 MPa) is 1.3 times lower then that (≈17 MPa) of the aged specimens. These values are also in good agreement with those of Butt and Khwaja [13] obtained with 99.996% Al polycrystals of six different mean grain-diameters in the range 0.25 to 0.51 mm. The observed 30% increase in yield stress due to room-temperature ageing may be ascribed to the migration of point defects, like vacancies and residual gaseous and metallic impurity atoms, to the cores of edge dislocations during the ageing period, which pinned the dislocations and made their movement rather difficult. Pinning of edge

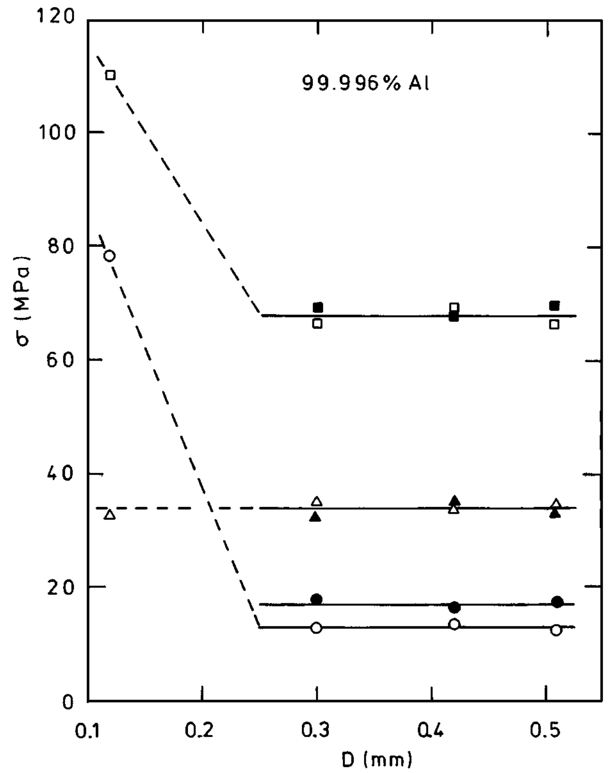

Figure 1 Dependence of yield stress (● ○), ultimate tensile stress (■ □) and fracture stress (▲△) of 500°C annealed 99.996% Al on the grain size. Filled and empty symbols refer to aged and un-aged Al, respectively. Data for un-annealed Al of 0.12 mm grain size have been given for comparison.

dislocations by hydrogen atoms diffused into 99.999% copper polycrystals, causing increase in the yield stress at rather low temperatures [15], supports this hypothesis.

On the other hand, there is no notable effect of ageing on the ultimate tensile stress and fracture stress of 99.996% Al polycrystals. The date points for ultimate tensile stress for both aged and un-aged specimens are very well encompassed by a straight line parallel to the D-axis at a stress level 68 MPa; same is the case for fracture stress but at a stress level 34 MPa. This is again consistent with the observations of Butt and Khwaja [13]. The nearly coincident values of ultimate tensile stress for aged and un-aged specimens indicate that pinning of glide dislocations by vacancies and residual impurity atoms, which is significantly enhanced during ageing period, becomes ineffective at substantially large strains. This proposition is substantiated by the investigations [e.g. 16–21] on the mode of deformation of metals and alloys using strain-rate cycling technique, which show that solute-atom pinning-points are effective obstacles to dislocation glide in the initial stage of plastic deformation in alloy crystals. However, on exceeding a certain value of plastic strain, dislocation-dislocation intersection becomes rate determining; resistance of the alloy crystals to plastic deformation then derives mainly from the forest cutting rather then from the solute-atom pinning-points.

It is also evident from Fig. 1 that yield stress and ultimate tensile stress of as-received 99.996% Al polycrystal ($D = 0.12$ mm) are 78 MPa and 110 MPa, respectively. These values are greater than the corresponding ones appertaining to annealed specimens (i.e. 13 MPa and 68 MPa) by a factor of 6 and 1.6, respectively. However, the fracture stress of as-received Al specimen is 32.5 MPa, which is in good agreement with that of annealed specimens. The substantial decrease in the values of yield stress and ultimate tensile stress of 99.996% Al polycrystals on annealing is a consequence of the reduction in the internal stresses and dislocation density in the annealed crystals. Akhtar *et al.* [22] have also observed that for a given grain size and a fixed stress level, the relaxation rate in Ti-6.5 at.% Nb polycrystals is significantly influenced by the annealing temperature; the higher the annealing temperature the lower is the relaxation rate because fewer mobile dislocations will be available to participate in the stress-relaxation process.

Now we shall dwell on the stress relaxation response of annealed 99.996% Al polycrystals. Stress-relaxation curves obtained with un-aged and aged Al specimens of grain size (a) 0.30 (b) 0.42 and (c) 0.51 mm are shown in Figs 2 and 3, respectively. For a given stress level σ_0 at which further deformation of the specimen was stopped, the amount of stress relaxed $\Delta\sigma(t) = \sigma_0 - \sigma(t)$ increases linearly with logarithm of relaxation time t, except for a few seconds in the beginning of the relaxation. These curves are therefore basically logarithmic, and comply with the relation

$$\Delta\sigma = \sigma_0 - \sigma = s \ln\left(1 + \frac{t}{t_0}\right) \tag{1}$$

Here σ is the 'relaxed' stress level at an instant of time t, $s = s(\sigma_0, T)$ and $t_0 = -s/\dot{\sigma}$ for $t \to 0$. The values of the slope

$$s = \frac{\mathrm{d}(\Delta\sigma)}{\mathrm{d}\ln\left(1 + \frac{t}{t_0}\right)} \tag{2}$$

as determined from the typical straight parts of $\Delta\sigma/\ln(t)$ curves, in which deviations from linearity occurred for initial few seconds due to the omission of t_0 in the conventional representation used, have been denoted by empty and filled symbols in Fig. 4 for un-aged and aged Al specimens, respectively. It is evident that relaxation rate s for a given initial stress level σ_0 is rather higher in the case of aged Al specimens as compared with that in un-aged ones. However, grain size has no effect on relaxtion rate in each case; the unit activation process of stress relaxation is therefore intragranular rather than intergranular.

The observed grain-size independence of the relaxation rate in coarse-grain materials has already been very well accounted for by Akhtar *et al.* [23]. Furthermore, the lines drawn through s/σ_0 data points for both types of Al specimens intersect the stress axis at $\sigma_i = 5$ MPa, which according to Roberts [24] is small, strain independent but temperature dependent, friction stress, or as termed by Hamersky and Trojanova [25], a threshold stress. The value of σ_i obtained in the present work is in good agreement with that obtained

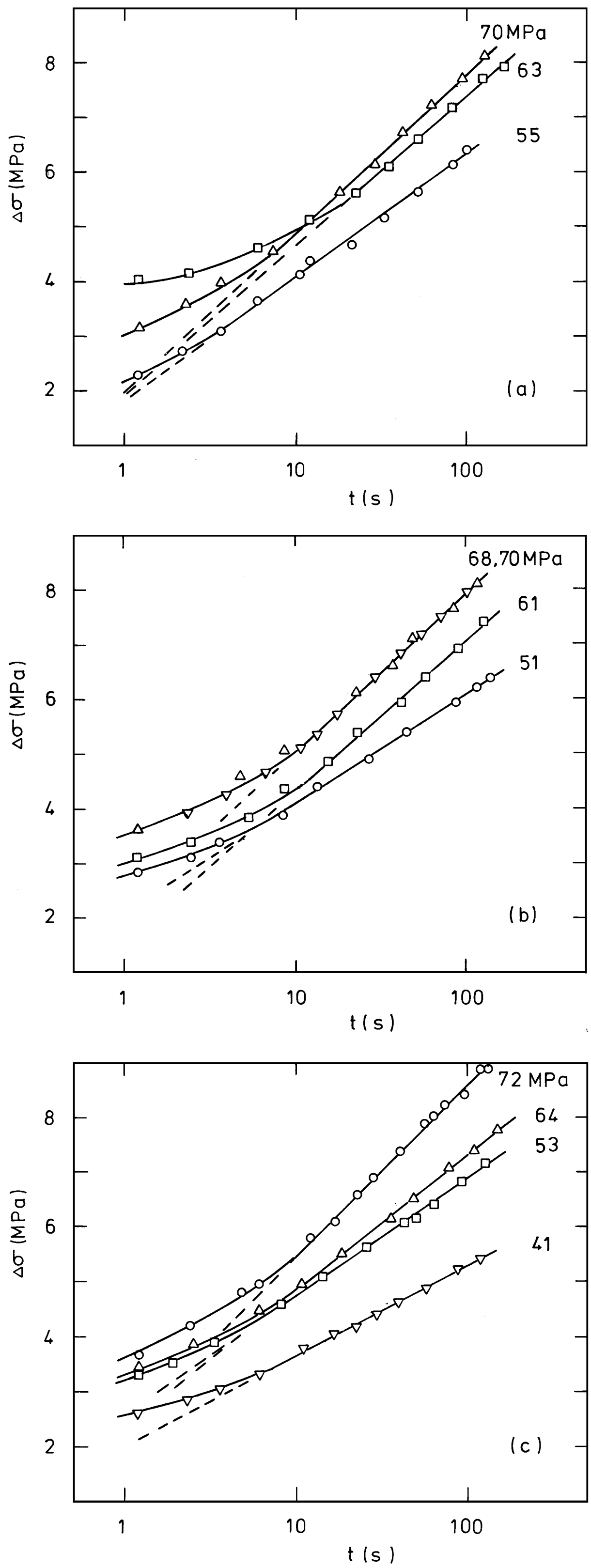

Figure 2 Stress relaxation curves obtained with 500°C annealed but un-aged Al of grain size (a) 0.30 (b) 0.42 and (c) 0.51 mm; $\Delta\sigma = \sigma_0 - \sigma(t)$, where σ_0 is the initial tensile stress denoted on the curves.

by strain-rate cycling experiments [14] carried out with the same material.

It is well known that activation volume v_σ associated with logarithmic stress-relaxation is given by [26]

$$v_\sigma = \frac{kT}{s} \tag{3}$$

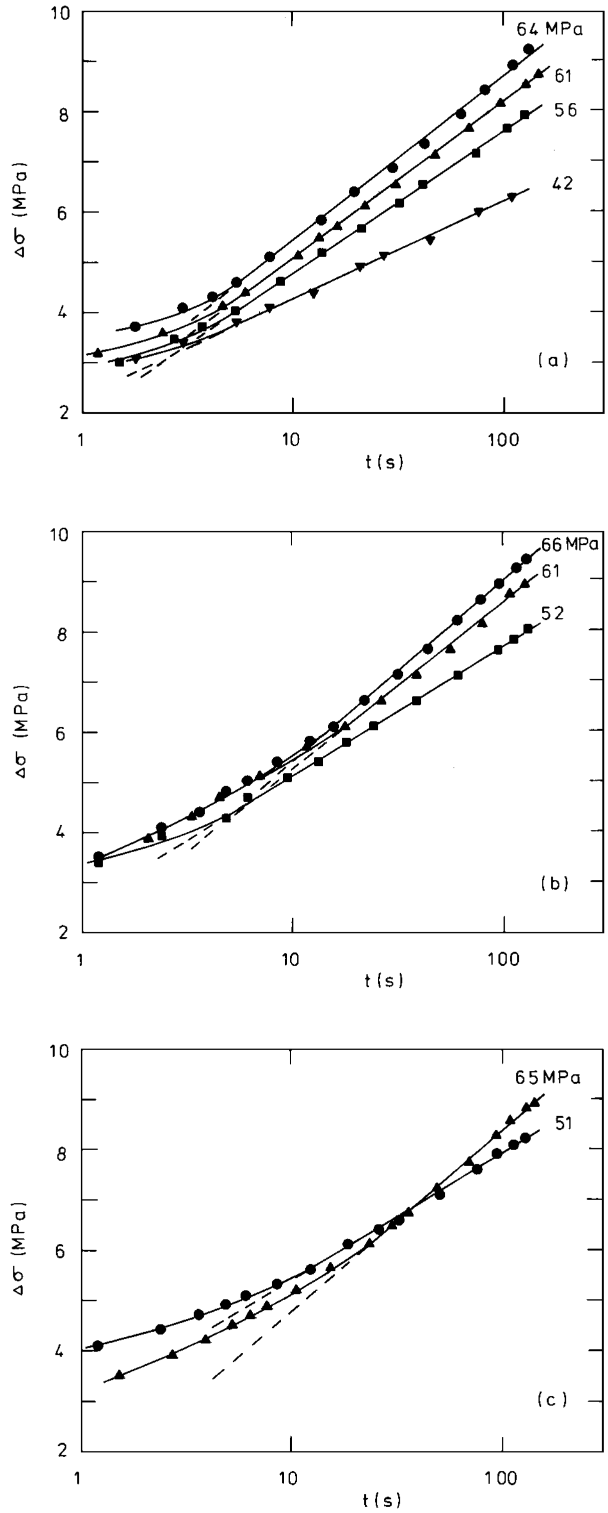

Figure 3 Same as Fig. 2 but for 500°C annealed and then room-temperature aged Al.

where k is the Boltzmann constant and T is the absolute temperature at which measurements are made. Using the s-values of Fig. 4, corresponding v_σ - values were calculated, and are depicted by symbols in Fig. 5 as a function of initial stress levels σ_0, and of $\sigma_0 - \sigma_i$, in double logarithmic coordinates. In each case, the data points for aged and un-aged Al specimens fall close to two separate but parallel lines. The slopes of these lines in the two cases are found to be $\mathrm{d}\ln v_\sigma/\mathrm{d}\ln\sigma_0 = -1.1$ and $\mathrm{d}\ln v_\sigma/\mathrm{d}\ln(\sigma_0 - \sigma_i) = -1.0$. This leads us to the

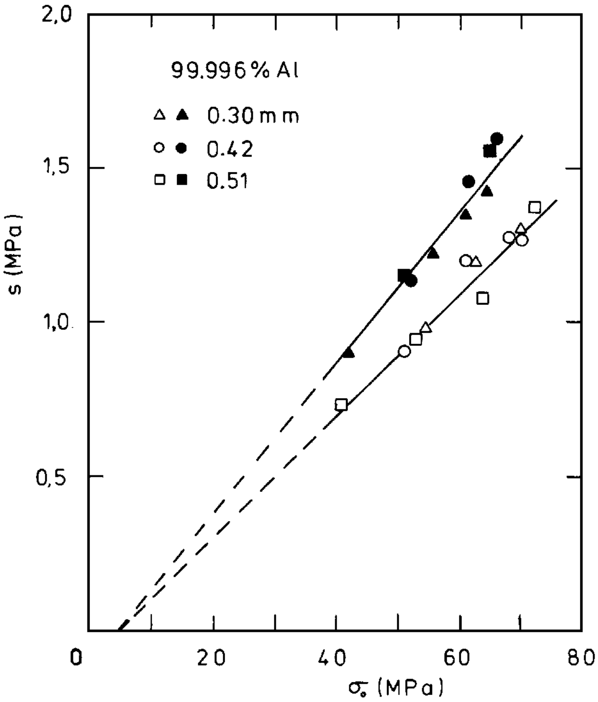

Figure 4 Stress-relaxation rate s as a function of initial stress level σ_0. Filled and empty symbols refer to aged and un-aged Al, respectively. Mean grain-diameters are (▲△) 0.30, (●○) 0.42 and (■□) 0.51 mm.

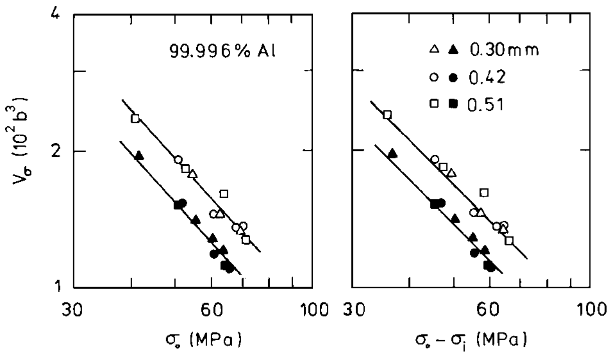

Figure 5 Activation volume v_σ as a function of initial stress level σ_0 and of $\sigma_0 - \sigma_i$ in double logarithmic representation. Notation is the same as in Fig. 4.

following mathematical expressions:

$$v_\sigma \sigma_0^{1.1}, C^* \tag{4}$$

$$v_\sigma(\sigma_0 - \sigma_i) = C \tag{5}$$

The value of constant C is 1.0 eV for aged specimens and 1.3 eV for un-aged specimens.

To elucidate the rate process of stress relaxation, we shall have recourse to a simple single barrier model of plastic flow [27, 28], as here embodied in the relation:

$$U_0 - v_\sigma \sigma = mkT \tag{6}$$

Here U_0 is the intrinsic height of the barrier governing dislocation movement, v_σ is the activation volume associated with stress σ, and $m \approx 25$. It should be noted that activation volume v_σ is also defined as $v_\sigma = (-\partial U/\partial\sigma)$, where $U(\sigma)$ is the activation energy necessary to overcome the barrier with aid of stress in the activation process or the "stress-reduced" barrier height such that $U(\sigma) = mkT$. Here $m = \ln(\dot{\varepsilon}_0/\dot{\varepsilon})$ is a constant which, on taking typical values of $\dot{\varepsilon}$ in the range 10^{-3} to 10^{-5} s^{-1} and with $\dot{\varepsilon}_0 \approx 10^7$ s^{-1}, comes out to be equal to about 25 [14, 28]. Replacing the term $v_\sigma \sigma$ in (6) by its 'equivalent' C from (5), the resulting modified expression yields on taking $m = 25$ and $T = 295$ K, $U_0 = 1.64$ eV for aged Al and $U_0 = 1.94$ eV for un-aged Al. These values are comparable with those found in the case of Ni (1.6 eV) [29], Co (1.6 eV) [26] and Ti (1.2 eV) [30] polycrystals. They are of the order of magnitude for recovery processes, e.g. cross slip, vacancy formation, mutual destruction of edge dislocations by forced climb in dipole configuration. It is interesting to note that the value of U_0 in Cu-Zn polycrystals [31] decreases by about 15% when the content of randomly distributed solute atoms is increased from 0 to 20% at.% Zn. Similarly, the migration of vacancies, residual gaseous and metallic impurity atoms to the cores of edge dislocations in aged Al polycrystals increases the concentration of point defects around the dislocations, which results in a decrease in the U_0 – value from 1.94 eV to 1.64 eV, i.e. by about 16%.

It should be noted that one can also get from (3) and (6) an expression for stress-relaxation rate s in terms of initial stress level σ_0, temperature T and intrinsic height of the thermally activable barrier controlling the dislocation motion U_0, which is as under [32, 33]:

$$s = \frac{kT\sigma_0}{(U_0 - mkT)} \tag{7}$$

This, in its differential form, can be re-written as

$$U_0 = kT\left[\left(\frac{\mathrm{d}\sigma_0}{\mathrm{d}s}\right) + m\right] \tag{8}$$

Now, from the slope of the lines drawn through the s/σ_0, data points in Fig. 4, one gets $\mathrm{d}\sigma_0/\mathrm{d}s$ equal to 40.9 and 50 MPa for aged and un-aged specimens, respectively. Substitution of these values in (8) leads to $U_0 = 1.67$ and 1.91 eV for aged and un-aged Al, which are identical with the U_0 – values already obtained by means of (5) and (6).

Reverting to Fig. 5, it is evident that for a given stress level σ_0, the assoicated activation volume v_σ is smaller than that in un-aged Al. This can be justified as follows. If L is the length of dislocation segment involved in the unit activation process, d is the average activation distance and b is the Burgers vector, then product Ldb will be the activation volume. Now, the value of activation volume (Ldb) will decrease if either L or d becomes small. It is established [19, 34] that values of L as well as d decrease as concentration of solute atoms in an alloy crystal increases. Thus, both L and d in aged Al will be smaller compared with the corresponding parameters in un-aged Al due to the enhanced concentration of point defects in the vicinity of edge dislocations. This accounts for the observation cited above.

4. Conclusions

We conclude from the foregoing evidence as follows.

1. The grain size has no notable effect on the strength parameters and stress-relaxation rate in both un-aged and aged high-purity aluminium.
2. The yield stress of annealed high-purity aluminium is increased by a factor of 1.3 on ageing it at room temperature for a long time. There is however no effect of ageing on ultimate tensile stress and fracture stress.
3. Room-temperature ageing of high-purity aluminium influences its stress-relaxation response. For a given stress level at which relaxation is allowed to start, the stress-relaxation rate in aged specimens increases by a factor of 1.3 while the associated activation volume decreases by the same factor as compared with that in un-aged specimens.
4. The intrinsic height of energy barrier to dislocation movement during stress relaxation at a constant strain in each case is of the order of magnitude for recovery processes.

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Note added in proof:
It is worthy of note that the theoretical expression (6) implies, on replacing σ by σ_0, $v_\sigma, \sigma_0 = C_0$ (constant), whereas experimentally it is found that v_σ varies with σ_0 such that $\sigma_0 = C^*/\sigma_0^{1.1}$ and $v_\sigma = C/(\sigma_0 - \sigma_i)$ as here embodied in (4) and (5), respectively. In spite of this inconsistency, it is still reasonable to infer values of $U_0 = 1.64$ eV for aged Al and $U_0 = 1.94$ eV for unaged Al from (6) in conjuncture with (5), as explained below. The experimental value of the product $v_\sigma \sigma_0$ for aged Al ranges from 1.17 to 1.10 eV with a mean value of $C_0 = 1.14$ eV, which is close to the corresponding value of $C = 1.0$ eV. Similarly, the measured value of the product $v_\sigma \sigma_0$ for unaged Al lies in the range 1.46 to 1.37 eV with a mean value of $C_0 = 1.42$ eV, which is again close to the corresponding value of $C = 1.3$ eV. Hence the values of U_0 referred to above, derived from (6) by using C values, will increase only by a rather small amount of 0.13 eV if one uses instead measured mean C_0 values. This will have no effect on the inference drawn above regarding the nature of the rate process of plastic flow in aluminium.