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MEDIUM TEMPERATURE PEAKS IN HIGH PURITY ALUMINIUM DOPED WITH 10 ppm COPPER OR SILVER

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Résumé : Les relaxations observées vers 0.5 Tm dans l'aluminium de pureté ordinaire ont des caractéristiques assez différentes de celles observées dans l'aluminium de très haute pureté. Dans cet article, les différences sont analysées à partir d'échantillons de quatre puretés différentes.

Summary : In the case of aluminium of ordinary purity, the observed relaxations around 0.5 Tm have a behaviour which is different from that of high purity aluminium. In this paper, the differences are analyzed using specimens of four different purities.

INTRODUCTION

The mechanisms which control plasticity and creep in metals around 0.5 Tm (Tm : melting point) were often in the center of interest for many experimenters. Recently numerous interesting works have described precisely several mechanisms. Before, proposed mechanisms around 0.5 Tm were cross-slip and self diffusion for the two stages U and U' of the creep in fcc metals /1/. Pipe-diffusion was considered in order to explain the stage U', but afterwards a cross-slip mechanism was generally accepted there.

Electron microscopy in-situ of the dislocations microstructure done in Toulouse /2,3,4/ has contributed to explain cross-slip mechanism controlling macroscopic creep in aluminium around 200C. This process can be activated only when the local stresses are high enough (about 0.75 of the Drowan stress). This value of the stress was measured in the Al-Zn alloy /5/. In fact such a process takes place after another mechanism, the glide in the non-compact slip plane {100}, by the kink nucleation at dislocation nodes /6/. The last mechanism has been recently analyzed by Carrard /7/, studying creep in aluminium monocrystals oriented <110>. Such creep in {100} plane can be responsible for the U' stage. Similar study was performed in magnesium and beryllium /8/ where the prismatic glide is observed at the analogous temperatures as in the case of {100} creep in aluminium.

For many years already, Internal Friction techniques have been used for studying metals in the similar temperature range /9,10/. In fcc metals of ordinary purity, a significant relaxation in polycrystalline materials was observed ("Ke peak"). The activation energy of that phenomenon is close to Hv (self diffusion energy). In monocrystals this relaxation is much less pronounced /11/. In all cases two additional and smaller peaks were observed /11,12/. The so called P1 peak of lower energy (.75 Hv) was present around 470K at 1 Hz.

In high purity aluminium the Ke peak was not observed /13/ and only the P1 peak exists in deformed samples. The P1 peak disappears consecutively after 10% tensile creep at 200C in stationary stage, and is replaced by another peak situated near the melting point /14,15/. The P1 peak can be re-developed by means of plastic deformation.

The aim of the present work is the study of the anelastic properties between .3 and .8 T_m in aluminium of different purities between zone refined 6N Al and ordinary 5N Al. We will try to answer the following question : Which of the relaxations in ordinary purity materials are related with the isolated dislocations, subboundaries or grain boundaries?

In this paper we will mainly expose the experimental results and their short discussion based on the already known models discussed in detail elsewhere /15,16/ and also in this conference in the paper headed "Internal Friction at Medium Temperatures in High Purity Aluminium" /17/.

EXPERIMENTAL

Samples have the form of plates 1x5x50 mm, prepared from polycrystalline aluminium annealed, deformed or after creep. The as-received state is the result of rolling an ingot ; a subsequent annealing up to 800K can produce the recrystallization. Four purities were used :

- Al 6N melted zone refined, elaborated by CECM Vitry/seine (France).
- Al 6N (previous) doped with 9.4 \pm 1 ppm of silver.
- Al 6N doped with 10,5 ppm \pm 1 ppm of copper.
- Al 5N with about 10 ppm of all impurities.

Except one of the 5N aluminium samples, the grain size was bigger than the thickness of the samples. Table 1 gives the grain size and the ratio of grain size to thickness of the samples.

Measurements were made at Genie Atomique Laboratory in the forced pendulum working under 1 torr of helium pressure. The measuring system consisted mainly of the 1250A Schlumberger Frequency Resoonse Analyzer equipped with appropriate excitation and detection transducers and amplifiers. The temperature regulator controlled by a HP71 pocket computer will be described elsewhere /18/. Data are transmitted to a HP9520 computer for further processing and archiving.

EXPERIMENTAL RESULTS

We have studied the internal friction behaviour as a function of parameters such as frequency of oscillation, oscillating stress amplitude, bias stress amplitude, microstructure (after coldwork or after creep in stationary state) on four samples of aluminium which possess different levels of purity and impurity content. In the case of 6N aluminium, the experimental results and their interpretation have been published already /15,16,19/ and will be developed in this conference in the companion paper previously mentioned. In this work we present the results concerning the aluminium with other purities.

Silver doped aluminium :

Fig. 1 presents the friction spectra of 6N aluminium doped with 10ppm silver for different frequencies of oscillation (in three decades) in the temperature range 0.3-0.8 T_m . Using a polynomial law, the internal friction background has been subtracted and results are presented in fig. 2. The activation energy of the observed peak has been obtained using the Arrhenius plot presented in fig.7 .The influence of the oscillating stress amplitude has been tested and a smooth decrease of the relaxation intensity is observed (factor of .9 in 3.5 E-6 - 3.5 E-5 range) /20/. The effect of a bias stress was also studied, a superimposed torsional stress was incremented at the maximum peak temperature of 545K. Absolutely no effect was detected /20/. The influence of the microstructural changes on the relaxation peak is shown in fig.3. The curve-a concerns aluminium in the as-

received state, the curve-b is obtained after a creep of the previous sample (8.3% tensile at 473K with a secondary stage creep rate of $5 \cdot 10^{-6} \text{ s}^{-1}$), the curve-c concerns the cold-worked sample after the previous creep (19% rolling at RT). As observed in 6N aluminium /14/, the creep leads to a disappearance of the relaxation peak and the coldwork provokes its reappearance with a significant decrease of internal friction in the high temperature range.

Copper doped aluminium :

The internal friction spectra of this sample are shown in fig.4 for different frequencies of oscillation. We observe a slightly higher maximum peak temperature as compared with the previous sample. Similar results as in the case of silver doped aluminium are obtained as a function of bias stress level at 560K, as a function of creep (the relaxation peak does not exist after 9% creep and a high internal friction increase takes place) and at last a well-defined relaxation is also detected as a function of coldwork after creep /20/.

The oscillating strain amplitude effect is slightly different, the peak height seems to keep a constant value in the $1\text{E}-6 - 5\text{E}-5$ range.

5N aluminium :

In this case two samples have been used. The first (1) (as-received and annealed) has a large grain size compared to its thickness. It presents a relaxation at 540K (1 Hz) (see fig.5, curve a) which was analyzed in three decades (fig.7). This relaxation slightly decreases with the oscillating strain amplitude (factor of .9 in the $3.5 \text{ E}-6 - 3.5 \text{ E}-5$ range). It is also independent of bias stress level at 540K /20/. The second sample (2) was crept 13% at 575K and had retained a small grain size (.3 mm compared with the thickness .8 mm). The results are shown in fig.5, curves b and c. Curve b gives the internal friction during first heating : no relaxation seems to be present at 540K but one is seen around 600K. It is followed by an important decrease at 700K which is correlated with an increase of modulus defect. Here a recrystallisation takes place leading to a grain size increase to 1.5 mm (hereafter called 5N aluminium (3)).

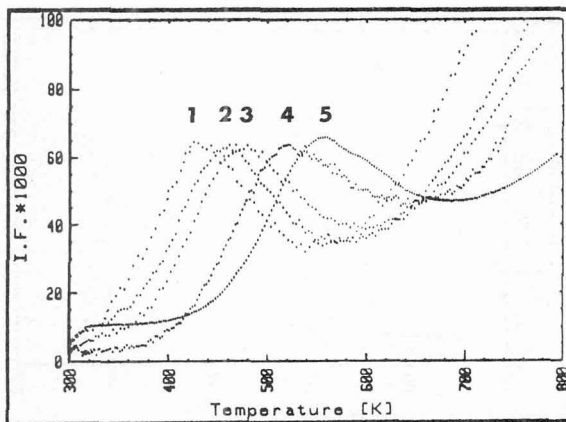


Fig 1 : 0-1 internal friction spectra of Ag doped Al for the frequencies : 1) .01 - 2) .05 - 3) .1 - 4) 1 - 5) 10 Hz (Oscillating ampl. $7\text{E}-6$ during cooling).

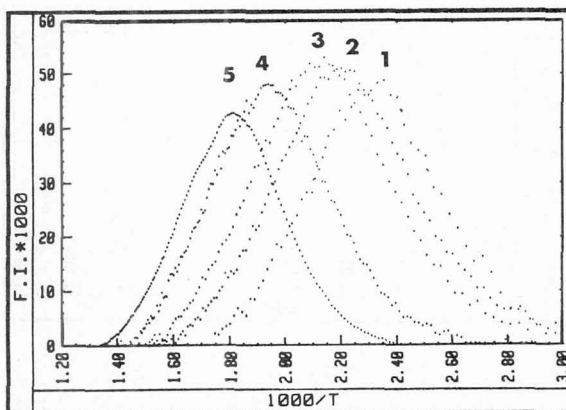


Fig. 2 : Same results as Fig. 1 with a subtraction of internal friction background.

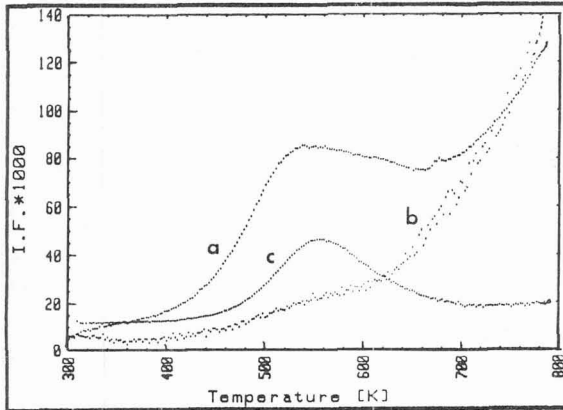


Fig. 3 : Ag doped Al spectra (Osc. ampl. $3.5 \text{ E-6} - 1 \text{ Hz}$)
 a) as-received and annealed 800K.
 b) as a + tensile creep of 8.3% at 473K.
 c) as b + coldwork by rolling of 19% at RT.

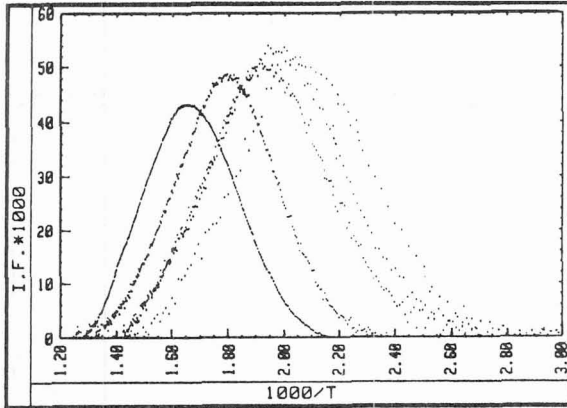


Fig. 4 : Cu doped Al spectra (Osc. ampl. 7 E-6 during cooling) without the internal friction background (from left to right : 10 - 0.1 - 0.05 - 0.01 Hz).

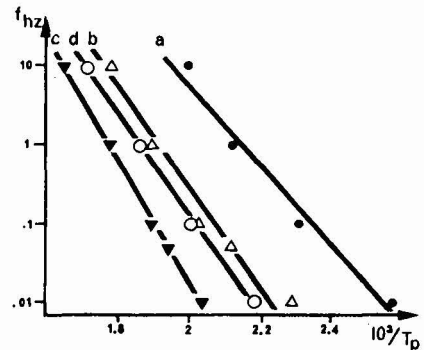


Fig. 7 : Arrhenius plots according to samples described in Fig. 6.
 a) 6N Al.
 b) Ag doped Al.
 c) Cu doped Al.
 d) 5N Al.

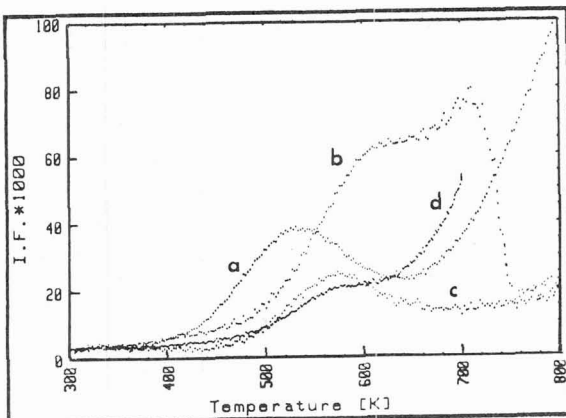


Fig. 5 : 5N spectra (Osc. ampl. : 7 E-6 , 1 Hz) :
 a) as-received and annealed at 800K (sample 1).
 b) not annealed and crept 13% at 575K (sample 2), first heating.
 c) as b + first cooling (sample 3)
 d) as c + tensile creep of 11% at 477K.

During cooling (see curve c) and subsequent heatings we observe a diminished relaxation at 560K. In order to test the behaviour of this last peak, we have performed a new creep experiment (11% creep at 477K -curve d). We see an important decrease of the relaxation as observed in our previous study on 6N doped samples having similar grain size.

Recapitulation :

In fig.6 we can compare internal friction spectra of aluminium with different levels of purity and also for similar grain size under the condition of grain size exceeding the sample thickness. Conclusions on the peak height difference are difficult because it is correlated with dislocation density before and during first annealing. The Arrhenius plots are shown in fig.7 and the best fits for activation energy and frequency factor are given in table 1.

Except in the case of 5N aluminium (2), the behaviour of all other samples is exactly the same, especially the disappearance of the relaxation after adequate creep. Also one observes a very deep evolution of

internal friction in the high temperature range : high after creep, low after coldwork and intermediate values after annealing of the as-received material.

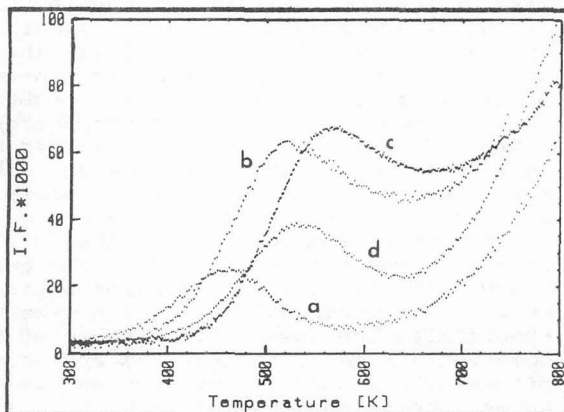


Fig. 6 : Comparison of internal friction spectra (osc. ampl. $7E-6$ during cooling, 1 Hz) for as-received and 800K annealed state : a) 6N Al - b) Ag doped Al - c) Cu doped Al - d) 5N Al (similar grain size around 1.5 μ m).

	Grain size \varnothing	Ratio \varnothing/e	Peak temp.	Activ. energy	Frequency factor f_0	After creep
Al 6N	1.5	1.5	470	1.01	1. E+11	No peak
Al Ag	2.	2.	526	1.27	1.7 E+12	No peak
Al Cu	1.5	1.5	562	1.56	9.7 E+13	No peak
Al 5N(1)	1.2	2.	537	1.27	8. E+11	---
Al 5N(2)	.3	.4	610	--	---	Peak
Al 5N(3)	1.5	1.8	570	--	---	No peak

Table 1 : Sample data (\varnothing grain size in mm, e thickness in mm, temperature in Kelvin, energy in eV and frequency factor in s⁻¹).

DISCUSSION

The more surprising result is the shift of the relaxation peak and change of the activation energy between the high purity sample and the others less pure. Undoubtedly, the activation for 6N aluminium cannot be compared with the bulk diffusion energy ($H_v \sim 1.3-1.5$ eV) and then the doubt exists for the other purities. Also we observe an increase of frequency factor in low purity samples which is compatible with a diminution of dislocation length L ($f_0 \sim 1/L^2$). Theoretically it is easy to explain a temperature shift in correlation with dislocation length evolution. But here the shift would be in a sense opposite to that observed. Hence the observed shift is mainly due to activation energy increase. Does this energy correlate with dislocation length

variation? Theoretically such a variation exists : $H=H_0-v\sigma$ but the variation of the activation volume v will be very large ($1.E4$ b3). In fact, such a large activation volume change will be easily detected by oscillating stress changes while not so large temperature shifts have been obtained ($< 20K$ between $3 E-6$ and $3 E-5 \mu$). We conclude that the observed relaxations are governed by one or more mechanisms with purely different characteristic energies. Nevertheless except for the case of the 5N aluminium (2) sample, the similarity in the behaviour of the relaxations of all other samples indicates that a common process of relaxation is involved. The intrinsic dislocation processes, such as cross-slip or non-compact $\{100\}$ glide, are not so sensitive to impurities. For the reasons explained in another paper of this conference /17/, the chosen process in 6N aluminium is the jog motion on screw dislocations governed by pipe-diffusion mechanism. Here it appears that impurities decrease the dislocation mobility which enables a more efficient process of vacancy nucleation in the bulk governed by the activation energy H_v to come into effect.

The relaxation observed in the 5N aluminium (2) sample on the contrary does not correspond to the previously described processes, since this relaxation is present after creep at 610K. So this affords us a direct observation of grain size effect. We can compare this result with the observations reported by the chinese group /21/ which observes different relaxations at the same temperatures. The existence of bamboo boundary in the specimen is a necessary condition for the appearance of the peak at 540K. Such a peak is attributed to the combined effect of the bamboo boundaries and the defect configuration as subboundaries. In fact the process giving the bamboo boundary peak will be known from microstructural characterizations by electron microscopic observations.

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