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Department of Metallurgical Engineering
Michigan Technological University
Houghton, Michigan

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SERRATED YIELDING IN SINGLE CRYSTALS OF SILVER-INDIUM ALLOYS

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BY Q.H. Kahn / E.J.T. amb
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Prepared by:

J. D. Johnson

A. A. Hendrickson

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SERRATED YIELDING IN SINGLE CRYSTALS
OF AG-IN ALLOYS

by J. D. Johnson and A. A. Hendrickson

ABSTRACT

Serrated yielding was observed in tensile tests of high purity single crystals of Ag-9.6 at/o In. The temperature dependence of the critical strain for the onset of serrations was measured. The estimated apparent activation energy is 0.6 to 1.0 ev. Techniques of preparing high purity and relatively strain free Ag-In single crystals of a selected orientation are discussed.

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INTRODUCTION

The Portevin-LeChatelier effect, commonly referred to as serrated yielding, has been reported to occur in a number of alloys: both fcc Al-Mg¹, Cu-Sn², Al-Cu³, and bcc Fe-N⁴ and Fe-C⁵. However, most of the information has been obtained from studies of polycrystalline materials which were frequently of only commercial purity. The uncertainties caused by the presence of grain boundaries and solid solutions of several solute elements has lead to a rather unclear picture as to the mechanism(s) controlling serrated flow. For these reasons it was decided to study in detail serrated yielding in high purity single crystals of one orientation. This report presents initial observations of serrated yielding in Ag-9.6 at. % In.

EXPERIMENTAL METHODS

Material

Silver and indium of 99.999% purity used in this investigation were obtained from the American Smelting and Refining Company. The alloy was prepared by placing the two components, weighted to the nearest gram of the 7 a/o composition desired, into a high purity vacuum degassed graphite crucible. The crucible and alloy constituents were inserted into a quartz tube, evacuated to about 1×10^{-5} mm Hg, purged and re-evacuated with high purity argon two times and finally sealed under 1/2 atmosphere of high purity argon.

Melting was accomplished by placing quartz tube assembly into a resistance wound tube furnace at 100°C . The alloy was kept molten for one hour at this temperature with periodic agitation being given to the quartz tube. The resulting ingot ($1/2'' \times 3/8'' \times 8''$) was etched in concentrated HNO_3 , cleaned in 5 parts NH_4OH , 3 parts 30% H_2O_2 , washed in distilled water and dried in high purity acetone, swagged in unlubricated dies to .250" rod which was sheared into 1" lengths. These were cleaned, encapsulated, and remelted as described above. The ingot was swagged to .218" and the resulting rod was cut into 5.25" lengths.

Single Crystal Seed Preparation

An initial seed of the desired orientation was prepared by placing pure silver in a 7 mm Vycor tube of which the lower portion had been bent into five 1" segments at about 45° to one another. This tube was placed in a 25 mm tube for support, sealed as before and lowered 9.0" into a resistance wound gradient

furnace containing a Pt baffle 9.5" below the top of the furnace at which point the temperature was maintained at $960.5 \pm .1^\circ\text{C}$. The whole assembly was held in position for one hour after the silver had melted and flowed into the bent portion of the tube to allow thermal equilibrium to be established. Actual growth was accomplished by lowering the support tube past the baffle at $.375"/\text{hr}$.

Upon cooling to room temperature the inner 7 mm tube shattered due to differential contraction and the fact that the tube was bent allowing a minimal amount of sliding between the tube wall and crystal to occur. Back reflection Laue patterns were made of the five portions to determine their rod axis. This procedure had to be repeated three times until a section of suitable orientation was produced. (See Figure 1.) The desired segment was spark cut from the remainder of the crystal and chemically machines in 50% HNO_3 to remove the deformation zone produced during spark cutting and reduce the diameter by about .5 mm.

The silver seed produced in the above manner was placed into a high purity vacuum degassed graphite mold, machines to fit loosely into a 25 mm Vycor tube. The mold was 8" long of which 6.0" were bored and reamed to .250" with the remaining 2.0" being left solid to increase the thermal gradient during crystal growth. Three inches of .218" diameter high purity silver rod was added to the mold which was then encapsulated and sealed under $1/2$ atmosphere of pure argon for increased thermal coupling.

The whole assembly was inserted into the gradient furnace to a depth which caused the top of the seed to be at the level of the baffle which was maintained at $960.5 \pm .1^\circ\text{C}$. A growth rate of

.375"/hr. was again used. Back reflection Laue photographs of the resulting single crystal were made to check orientation and for the presence of striation boundaries. Finding no striation boundaries and an identical orientation to the original seed, three 3/4" lengths were spark cut and three additional seed crystals were grown as described above.

Alloy Single Crystal Preparation

A seed section along with a 5.25" length of alloy rod were cleaned, placed into a mold and encapsulated as above. This assembly was lowered into the gradient furnace to a depth such that the top of the seed was level with the baffle which was now maintained at $900 \pm .1^\circ\text{C}$, held in position for two hours then lowered at .250"/hr. This reduced growth rate, over that used for seed production, was found necessary in order to insure single crystals having an absence of striation boundaries.

Upon completion of growth the mold assembly was removed from the furnace and allowed to cool in air. The quartz tube was opened and the rod extracted. In all cases it was found to slide freely from the graphite mold. Each rod was given a light etch in 10% HNO_3 to insure its single crystal nature after which the seed along with 2.3" of the lower portion of the crystal were spark-cut off. Cut surfaces were chemically machines to remove spark cutting damage in 50% HNO_3 . At this point the seed was ready for reuse. Back reflection Laues were made to determine the exact orientation of each alloy crystal section where upon they were individually sealed in small Vycor tubes under a vacuum of 2×10^{-6} mm Hg and annealed for 2 weeks at $800 \pm 5^\circ\text{C}$ in order to reduce coring.

Since considerable In redistribution was expected to occur during growth, an alloy crystal was sectioned every 1/2" and analyzed for In. The concentration of In as a function of distance from the seed is shown in Figure 2. Although the In concentration is seen to differ considerably between ends of the crystal the initial plateau from which the specimens were cut is a quite uniform 9.6 a/o.

Tensile Specimen Preparation

Tensile specimens having a 19 mm gauge section were spark turned using brass electrodes and a kerosene electrolyte from the lower portion of each crystal (Fig. 2). Thirty degree conical ends were provided for holding in split cup grips during tensile testing (Fig. 3). The gauge diameter was monitored with micro-meters down to 3.5 mm during rough spark cutting. The last two stages however, were carried out with no measurements for one hour each, resulting in a final gauge diameter of about 3.1 mm which was measured optically. Variation in section diameter from end to end at various rotations was found in all cases to be less than .05 mm.

Final surfaces were prepared by chemically polishing in a solution of 10 parts saturated chromic acid, 1 part HCl, 1 part H₂SO₄ and 10 parts H₂O until a bright mirror finish was obtained. The finished tensile specimen was then cut free of the remaining crystal and the cut surface was carefully chemically machines to prevent recrystallization during annealing. Each specimen was placed in a graphite crucible and annealed under a vacuum of 1×10^{-6} mm Hg at $700 \pm 5^\circ\text{C}$ for 24 hours*. After annealing no evidence

*Higher temperatures resulted in thermal etching.

could be detected of spark machining damage by means of x-ray diffraction

Tensile Testing

Crystals were tested on a flow model Instron tensile testing machine equipped with stepped zero suppression allowing a load sensitivity of $\pm .05$ lb. at all times. Specimens were held in precision split cup grips and placed on the tensile machine with the aid of a mounting jig (Fig. 4). Ball and socket swivels were provided at each end of the grips to ensure axial alignment.

Testing temperatures varied between 27°C and 100°C and were obtained by use of an environmental chamber and a precision resistance furnace controlled with an L&N AZAR Controller equipped with a C.A.T. unit driving a magnetic amplifier. Purified argon at the rate of $20 \text{ ft}^3/\text{hour}$ was used to maintain a positive pressure in the chamber during testing. A thermocouple was placed at the initial level of the gauge length center very close to the specimen (Fig. 5). Another thermocouple close to the furnace element was used for control. To date only testing at a strain rate of $4.4 \times 10^{-5} \text{ sec}^{-1}$ has been used.

RESULTS AND DISCUSSION

A typical flow curve, omitting serrations, for the Ag-9.6 a/o In alloy is shown in Figure 6. The small, rather broad yield point could be observed to 75°C; below this temperature serrations were of such a magnitude and occurred at such small strains as to make the presence of a yield point uncertain.

Within the usual range of values quoted in the literature⁶⁻¹⁰ of 1.5 - 2.5 for $(m+n)$, the sum of the exponents describing the rates of vacancy production and dislocation multiplication, the measured apparent activation energy is .60 ev to 1.00 ev. Since the migration energy of a single vacancy in Ag is approximately .7 ev¹¹, it is not possible at this time to even speculate as to the possibility of a solute lowered vacancy migration energy. The experimental value of $(m+n)$ is currently being determined from measurement of the strain rate dependence of the critical strain (ϵ_p). However, since the temperature dependence of ϵ_p is linear even at moderate temperatures it would appear unnecessary to consider thermal vacancies in the theoretical structure of the analysis.

A normal stress relaxation curve is shown in Fig. 8. However, under conditions where logarithmic creep occurred, a strong yield point on restraining was also observed. Figure 9 shows a curve of yield point return and is in excellent agreement with theoretically predicted slope of .67. At longer times the rate of increase of $\Delta\tau$ is much smaller indicating saturation of the solute atmosphere.

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Fig. 1 Orientation of seed and alloy single crystals.

Fig. 2 In distribution in alloy single crystal.

Fig. 3 Tensile specimens before (bottom) and after (top) testing.

Fig. 4 Tensile grips and mounting jig.

Fig. 5 Specimen in place on tensile machine showing control and monitoring thermocouples.

Fig. 6 Stress-strain curve of Ag-9.6 a/o In at 35°C.

Fig. 7 Temperature dependence of critical strain.

Fig. 8 Stress Relaxation curve.

Fig. 9 Yield point return curve.