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A PROGRAM OF RESEARCH ON MECHANICAL METALLURGY
AS RELATED TO FUEL-ELEMENT FABRICATION

QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING JUNE 30, 1961

Contract AT(04-3)-167
Project Agreement No. 4
U. S. Atomic Energy Commission

August 31, 1961

395 001

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GA-2502

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August 31, 1961

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I. DEFORMATION AND FABRICATION CHARACTERISTICS OF REACTOR MATERIALS

ROD DRAWING

The results of tensile tests performed on tube and rod specimens obtained from drawn bars were presented in the previous quarterly report.^{(1)*} The anomalous behavior observed in these tests was attributed to a radial variation in grain size of the starting material, which was of larger diameter than that normally used for the drawing experiments. Large-diameter rod was initially used to ease the problem of fabricating tube specimens; however, the results obtained were not useful for comparison purposes.

In view of the difficulties encountered, subsequent drawing experiments were performed on normal (3/8-in. initial diameter) bars, and tubular and rod test specimens were obtained from these drawn bars. The method of specimen preparation was previously described.⁽¹⁾

The results of these tests are shown in Fig. 1.1, in which the yield stress for each specimen is plotted versus the true strain experienced in drawing. As can be seen, there is as much as a 10% difference in yield stress between the inner rod specimen and the outermost tube specimen for $\alpha = 3^\circ$, $\epsilon = 0.535$. Table 1 lists the average flow stress for each reduction obtained by weighting each individual flow stress and area as described previously.⁽¹⁾

A comparison of the average computed yield stress for these tests with the previously obtained corrected master stress-strain curve is made in Fig. 1.2. It can be seen that the average yield stress obtained for $\alpha = 3^\circ$ is always greater than the stress for the same strain in simple

*References are listed at the end of each section.

Fig. 1.1--Yield-stress curves for tubular and rod specimens following drawing to the strain levels shown

Fig. 1.2--Comparison of average computed yield stresses for $\alpha = 3^\circ$ and $\alpha = 15^\circ$ with master curve

Table 1
RESULTS OF TUBE AND ROD TESTS ON DRAWN BAR

$\alpha = 3^\circ$		$\alpha = 15^\circ$	
True Strain, ϵ (in./in.)	Computed Avg. Stress (psi)	True Strain, ϵ (in./in.)	Computed Avg. Stress (psi)
0.088	12,340	0.153	15,080
0.535	18,420	0.556	18,600

tension. These results are quite different from those obtained by simply comparing the yield stress of drawn bars with the master curve,⁽²⁾ and indeed point out the importance of residual stresses in determining average true yield stresses. The previously obtained envelope of tensile yield stress following drawing through a die semiangle, $\alpha = 3^\circ$ over the range of strains shown is included for comparative purposes.

The contention of previous investigators⁽²⁾⁽³⁾ that a reasonably accurate and complete true stress-true strain curve beyond necking is provided by the envelope of a series of yield-stress curves having their origin on the strain axis at the point corresponding to the strain experienced in drawing, must be questioned in view of these results. Figure 1.3, obtained from grid-line experiments, also indicates the occurrence of shearing for bars drawn through a die semiangle of 3° and the reductions shown. Figure 1.3 lends qualitative support to the results obtained from tube tests.

The calculation of nonhomogeneous work by curve matching with the master curve for $\alpha = 15^\circ$ (Fig. 1.2), yields values lower than those obtained mainly from theoretical considerations.⁽⁴⁾ In view of the limited number of tube tests performed, this neither supports nor refutes the previous work.

A topical report covering the rod-drawing investigation will be prepared.

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$$\alpha = 3^{\circ}, \epsilon = 0.151$$

$$\alpha = 3^{\circ}, \epsilon = 0.580$$

Fig. 1.3--Grid-line specimens drawn to true-strain values as shown

EXTENSION OF MECHANICAL METALLURGY INVESTIGATION TO BCC METALS

One of the most serious problems in the area of formability of reactor materials is the generally brittle nature near room temperature of some high-temperature materials, such as the refractory bcc metals. Coupled with this problem is the tendency of such metals to exhibit an enhancement of brittleness under certain conditions in a radiation field. Since it is recognized that future reactor materials will be called upon to work at even higher temperatures and under more severe radiation conditions, it is clear that the problems presented by the refractory bcc metals point up the basic formability problem inherent in the general class of these materials--the ductile-to-brittle transition. This problem can be stated as: What determines the transition from a mode of deformation involving the generation and propagation of dislocations, which results in plastic deformation, to a mode of deformation involving the generation and propagation of microcracks, which results in cleavage and brittle fracture?

It is believed that there are several mechanisms that restrict the plastic mode of deformation in the refractory bcc metals, and this will determine when the ductile-to-brittle transition will occur. These mechanisms are

1. The inherently strong interaction potential between bcc dislocations and interstitial defects, which are common contaminants in the metals and can restrict both the mobility and the generation of dislocations.
2. The relatively strong Peierls potentials in the refractory bcc metals, which imply a rather strong interaction of the dislocation with its matrix lattice, so that this interaction can modify the dislocation mobility.
3. The strong temperature dependence of the retarding forces acting on dislocations in the refractory bcc lattices, especially as the temperature is lowered, which can be expected to result from (1) and (2).

4. The strong influence on the dislocation mobility, and hence on the plastic properties, of the rate of application of stress, as implied by several recent models of dislocations. (5)(6)

A portion of the program in mechanical metallurgy (the relation of formability to imperfection structure) has been devoted to a study of the bcc metals and more specifically to the study of dislocation interactions by means of stress-relaxation methods and amplitude-dependent internal-friction methods. From this phase of the program the interaction potentials of dislocations with their lattice, more commonly known as Peierls potentials, have been measured. From these measurements several important contributions have already been made: (1) The Peierls potential for dislocations in four bcc refractory metals--niobium, molybdenum, tantalum, and tungsten--has been measured; (2) it has been shown that dislocations in the bcc lattice are not free to glide without thermal activation at low temperatures; and (3) the measurements have separated impurity interactions with dislocations from intrinsic lattice interactions in ultrapure tantalum. Indeed, a portion of the program is presently devoted to obtaining ultrapure bcc metals after the method described by Seraphim, et al. (7) Tantalum having a residual resistivity ratio, Γ , of 1300 has been obtained in this way. Internal-friction measurements are continuing on this material. It is necessary and desirable to extend the range of measurements to lower frequencies where more information can be obtained about the internal-friction temperature spectrum and the effects of cold work on it. In addition, it is highly desirable to perform mechanical property measurements on this class of metals, using samples in which the interactions of impurities with dislocations have been reduced to a minimum by virtue of material purity. Of considerable importance are (1) the evaluation of theoretical models in order to determine the mechanisms controlling the flow of these ultrapure bcc metals; (2) an understanding of the nature of the frictional stress which seems to be common to all bcc metals; (3) an explanation of the yield point in bcc metals and some knowledge concerning the phenomenon

of the ductile-brittle transformation; and (4) the exact effect of defects, including those due to irradiation, on the lattice.

Measurements on ultrapure bcc metals followed by similar measurements in which the metals have been inoculated with known amounts of interstitial impurities such as nitrogen, oxygen, and carbon, and, ultimately, investigation of radiation-induced defects, should provide considerable insight into the manner in which point defects interact with and affect the behavior of the bcc metals. To this end, we plan to perform the following types of experiments in this program:

1. To extend the frequency range of the internal-friction measurements to very low frequencies (typically 0.005 c/s).
2. To determine the mechanical properties of the specially prepared bcc metals.

The present work has been concentrated in two areas: first, the building and testing of a microextensometer; second, the design and building of an electron-beam floating-zone furnace to operate at pressures of approximately 10^{-10} mm Hg. The floating-zone furnace will aid considerably in the attainment of ultrapure starting materials for this program.

MICROEXTENSOMETRY

A microextensometer designed to fulfill the following conditions has been designed and built and is in the process of being tested. First, the extensometer should be capable of operating over the temperature range 78° to $\sim 800^{\circ}$ K. Second, it should be capable of testing wire specimens and maintain good axial alignment during the testing of these specimens in tension. Third, it should be capable of measuring microstrains; that is, it should have a resolution of at least 1×10^{-6} . Figure 1.4a is a drawing of the extensometer which has been constructed; Fig. 1.4b presents a trace of the noise level of the system with zero load at room temperature.

Fig. 1. 4--(a) Drawing and schematic of microextensometry and system;
 (b) electrical noise level versus time for microextensometer and associated electrical system

As can be noted from Fig. 1.4b, the electrical noise level is approximately 1×10^{-7} .

Evaluation tests are planned soon for the over-all system.

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II. THE STORED ENERGY OF COLD WORK

The investigations of the influence of grain size on the stored energy of cold work have been extended to a new lot of OFHC copper. The results were consistent with the earlier work; the fine-grain specimens store more energy than the coarse-grain specimens and the stored-energy-release spectra differ in a manner similar to that observed earlier. However, there are some significant differences in the quantities of stored energy and the temperatures of recrystallization; these differences are tentatively ascribed to a strong sensitivity of the energy storage and release characteristics to the impurities present in the copper.

Stored-energy measurements were conducted on a new lot of OFHC copper, designated henceforth as Lot B. The experimental conditions of deformation and stored-energy release were identical to those employed earlier with Lot A; ⁽¹⁾⁽²⁾ the deformation was made in compression at 0°C, and the stored-energy release was conducted with a nominal heating rate of 4°C min⁻¹. A comparison of the results for the two lots of copper shows that the copper of Lot B (1) stores more energy, (2) recrystallizes at lower temperatures, and (3) yields a sharper energy release.

The results obtained to date for Lot B are summarized in Table 2.1. These results are preliminary in nature, since further work is currently in progress to resolve some effects which may be artifacts in the base-line heating curves. The specimens of different grain size were prepared by the methods described previously, ⁽¹⁾ but after making improvements in the deformation apparatus to retain alignment during the cubic compressions of 25%. These improvements have resulted in the attainment of more uniform grain size throughout each copper block. However, it was found that the coarse-grain specimens possessed an average grain size of only

Table 2.1

MEASUREMENTS OF STORED ENERGY IN OFHC COPPER, LOT B

Experiment No.	True Strain in Compression	Deformation Work, E_w (cal g-atom ⁻¹)	Stored Energy, E_s (cal g-atom ⁻¹)	Temp. of Release Peak, T_r (°C)	Ratio of Stored to Expended Energy, E_s/E_w (%)
Fine Grain (Avg. Grain Size, 20 μ)					
81	-0.131	36.6	2.70	371	7.38
82	-0.145	39.6	2.70	361	6.72
77	-0.231	78.4	5.08	327	6.32
76	-0.245	77.6	5.68	322	7.32
83	-0.307	110.7	7.45	300	6.23
84	-0.350	138.2	8.97	289	6.48
74	-0.392	163.0	8.31	280	5.10
75	-0.402	172.5	7.81	278	4.52
79	-0.670	342.5	8.65	234	2.52
80	-0.691	349.0	----	234	----
Coarse Grain (Avg. Grain Size, 50 μ)					
89	-0.243	-----	4.50	332	6.17
87	-0.312	115.0	6.25	310	5.43
85	-0.317	118.0	6.58	307	5.58
90	-0.368	151.6	7.58	297	5.00
86	-0.383	154.5	----	292	----
91	-0.546	216.0	9.29	263	4.30
88	-0.552	219.0	8.27	264	3.77

50 μ ; for comparison, the grain size of the Lot A specimens given similar treatment was 150 μ . In further work, studies will be made of the compressions required to obtain coarser-grained specimens with Lot B copper.

The total stored energies as a function of true strain are shown in Fig. 2.1 for specimens of average grain sizes of 20 and 50 μ . Some of the scatter in the data is believed to result from including all apparent stored-energy release in the integration to obtain E_s ; this includes a long, flat apparent release just prior to the recrystallization peak. This flat release was not satisfactorily reproducible in every case and was not observed in the measurements on Lot A. Furthermore, a broad peak of relatively small total area was frequently observed at temperatures near 425°C. No explanation is offered for these effects at this time. However, it is conceivable that the segregation and diffusion of impurities after recrystallization could be responsible; the estimated purity of Lot B is 99.990 wt-% copper, which is somewhat lower than the value of 99.993 wt-% copper previously estimated from the chemical analysis of Lot A.

The results shown in Fig. 2.1 are in general agreement with the data of Clarebrough, et al.,⁽³⁾ discussed in a previous report.⁽²⁾ The present results appear to give further definition to the functional dependence of the stored energy on the true strain, and the investigations will be continued with emphasis on the region of high strain, where there is some suggestion of a saturation in total stored energy. It should also be noted that satisfactory results have been obtained of true strains as low as -0.13.

The recrystallization temperatures, defined for the present purpose as the peak in the stored-energy-release curve, are shown as a function of true strain in Fig. 2.2. Despite the small difference in grain size, the difference in recrystallization temperatures for the two sets of specimens is well defined. A comparison of the results for the fine-grain specimens of Lots A and B (average grain diameters of 20 μ in both cases) shows that the recrystallization temperatures for Lot A were generally 40° to 60°C higher than for Lot B.

Fig. 2.1--Stored energy of cold work in OFHC copper deformed in compression at 0°C plotted as a function of true strain; results are for Lot B with average grain diameters of 20 and 50 μ

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Fig. 2.2--Effect of grain size prior to deformation on the recrystallization temperature of OFHC copper, Lot B, as a function of true strain; average grain diameters were 20 and 50 μ

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III. THE RELATION OF FORMABILITY TO IMPERFECTION STRUCTURE

It has been shown in previous reports and publications resulting from the present investigation⁽¹⁻⁴⁾ that the activation energy for motion of dislocation segments in plastically deformed bcc metals such as niobium and molybdenum can be measured by means of a study of their internal-friction relaxation spectra. It has also been shown that in these metals there exists a distribution of activation energies which is roughly divided into two broad energy bands corresponding to the α and β relaxation peaks. Furthermore, it has been shown that the number of dislocation segments within a given energy band changes with increasing amounts of prestrain. Thus, a study of the relaxation spectra of these metals can provide information concerning the dislocation structure following plastic deformation.

During these preliminary studies, it became clear that small amounts of interstitial impurities were significantly modifying the relaxation spectra. This was particularly evident in the low-temperature annealing phenomenon later identified as strain aging in molybdenum (see Ref. 2, p. 113). The impurity effects were also evident in the relaxation spectrum of zone-refined molybdenum, as reported in Ref. 1. In that study it was discovered that as the interstitial impurity level decreased, the α and β peaks of molybdenum began to lose their separation. This peak modification with increased purity was interpreted to mean that an internal-friction background component increased in strength relative to the relaxation component as the number of interstitial impurities decreased.

The recent availability of the very pure tantalum specimen, Ta-6, made possible a detailed study of the internal-friction relaxation spectrum relatively unmodified by impurities. Figure 3.1 shows the relaxation spectra, measured at 6 cps, of specimens Ta-7 and Ta-8 as deformed 4% and 12%,

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Fig. 3.1--Internal-friction spectrum of impure tantalum prestrained various amounts

respectively, in torsion. Specimen Ta-7 was NRC 40-mil stock as-received except for a preanneal of 1 hr at 950°C ; its ratio of room-temperature resistance to 5°K resistance, Γ , was about 8. Specimen Ta-8 was NRC 40-mil stock also as-received except for a preanneal of 10 min at 1500°C ; its Γ ratio was also about 8. This specimen was recrystallized. It can be seen that in both of these relatively impure samples the α peak is evident at 120°K , while the β peak begins at 180°K and extends up to the vicinity of 220°K . It is also clear from Fig. 3.1 that neither of the peak heights exceed 100×10^{-5} for a 12% prestrain.

In contrast to the above relatively well defined set of peaks, Fig. 3.2 shows the broad internal-friction spectrum found in specimen Ta-6. It can be seen that both the α and β peaks are evident in this pure sample but are considerably modified. The most obvious change is the addition of a background component which fills the region between the two peaks. Also, this background is manifested by the failure of the internal friction to descend on the high-temperature side of the β peaks.

It can be seen for a 14% prestrain that even if an estimated background component be subtracted from the spectrum, the α and β peaks are considerably higher than the corresponding peaks found in the less pure specimens Ta-7 and Ta-8 prestrained 12%. Thus, it appears that the impurities contained in specimens Ta-7 and Ta-8 tend to reduce the relaxation strength as well as to reduce the background component.

It is particularly interesting that as the amount of deformation increases beyond 15%, the relaxation component continues to rise, as is evident in the 53%-prestrained case. The peaks have merged and have risen to the very large peak-height value of 500×10^{-5} .

Of particular interest is the suppression of the background component on the high-temperature side of the β peaks for the highly deformed specimen. Evidently, either the point defects produced by the deformation or the dislocation-dislocation interaction has acted to suppress the background component for this amount of prestrain.

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Fig. 3.2--Internal-friction spectrum of pure tantalum prestrained various amounts

The following, then, is a short summary of the effects of interstitial impurities on the internal-friction spectrum of tantalum:

1. The broad background component first seen in zone-refined molybdenum and later seen in very pure tantalum is reduced, leaving only the relaxation component.
2. The peak height for a given amount of prestrain is lower in the impure specimens.
3. The maximum in the peak height versus amount of prestrain curve occurs at smaller amounts of prestrain, and the height of the maximum is reduced for increased amounts of impurities.

Since small amounts of interstitial impurities are known to play an important role in the mechanical properties of the refractory bcc metals, it is very interesting to find such strong impurity effects on the internal-friction relaxation spectrum. A study is now under way to investigate the frequency dependence of the background component in the very pure samples to determine whether it is thermally activated or obeys the quasi-viscous damping law predicted by Lothe. (5)(6)

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