

EFFECTS OF HYDROSTATIC PRESSURE
ON THE MECHANICAL BEHAVIOR OF
BODY CENTERED CUBIC REFRACTORY METALS AND ALLOYS

(NASA Research Grant No. NsG-654)

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Department of Metallurgy
Case Institute of Technology
Cleveland, Ohio

ABSTRACT

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Measurements of the influence of hydrostatic pressures up to 25 kilobars on the terminal ductile-brittle transition temperature of powder metallurgy tungsten wire of commercial purity have been carried out for annealing temperatures of 1310°C and 1600°C. The transition temperatures (80°C and 190°C respectively) for these vacuum annealed conditions are unaffected, within the accuracy of measuring T_d , by pressure cycling. In contrast, previous results for a higher annealing temperature, giving a coarser grained structure, showed a depression in T_d of the order of 50°C. The role of the perfection of substructure in determining the influence of pressure is being investigated further. Analogous observations for a tungsten-1% thoria alloy vacuum annealed at 2000°C and 2200°C showed no measurable change in T_d (130°C and 180°C respectively) on pressure cycling. The optical microstructures indicate that, even at 2200°C, the fiber structure is not completely removed. As for the tungsten, higher annealing temperatures are being used in further investigations and thin-foil observations made of the substructures developed.

In the case of the model system iron-carbon suitable methods of specimen preparation and observation of yield behavior in tension have been established for large specimens (0.15 in gage diameter x 1 in. gage length) from which thin foil specimens can be prepared directly. Tests on annealed specimens of a Fe-0.065%C alloy cycled up to 20 kilobars show the expected successive lowering of the stress for yielding and the elimination of the upper yield point, and confirm the selection of this particular model system. Further observations of pressure effects on yield behavior are being made, especially for the micro-yield range, and related to associated changes in substructure.

Author

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1. INTRODUCTION

Promising possibilities for improving the mechanical behavior of the refractory metals - in particular, those in Group VIA - have now arisen as the result of observations of the effects of high hydrostatic pressure on the mechanical behavior of metals and alloys. The phenomena of interest consist in (a) effects on terminal behavior, that is on the properties measured at atmospheric pressure after the application of a high pressure cycle^(1,2); and (b) effects of enhancing plasticity of solid material at high pressure⁽³⁾. The latter is currently being exploited in such processes as hydrostatic or ramless extrusion⁽⁴⁾ and shows considerable promise for the cold shaping of materials which can normally be worked only at elevated temperature or not at all, and for the improvement of extrusion efficiency, die life and the uniformity of cold-working throughout the material extruded. Applications of hydrostatic forming to production processes have already been reported⁽⁵⁾. Hydrostatic pressure cycling differs in that, while no shape change occurs in the metal subjected to the cycle, for certain metals the terminal mechanical properties exhibit changes such as to facilitate subsequent plastic deformation.

The present research program on the effects of hydrostatic pressure on the structure and mechanical behavior of body centered cubic metals (NASA Grant No. NsG-654) was begun on June 1, 1964, with the initial objectives of (a) testing the dislocation generation model proposed to account for the substantial decrease in the yield stress of iron carbon alloys observed after pressure cycling; and (b) investigating the response of tungsten and tungsten-thoria alloys to such pressure treatment.

The studies at Case during the first year of the present research program ^(6,7) showed by transmission electron microscopy observation that new dislocations are developed at second phase particles in iron and iron-carbon alloys as the result of the application of hydrostatic pressures in the range from 5 to 20 kilobars at room temperature. These results, which have since been published ⁽⁸⁾, support the hypothesis that the changes in yield behavior found in iron after subjection to high hydrostatic pressures arise from the generation of mobile dislocations by the localized stress concentrations developed at the interfaces between second phase particles and the matrix as a consequence of the different linear compressibilities of the two phases. However, the results also show that the nature and arrangement of the dislocation arrays which form is more complex - especially for the higher pressures and larger particles - than had been expected. The precise influence of particle type and morphology, in addition to the magnitude of the peak pressure, in determining this complexity is thus far from clear. As part of the effort to develop to a quantitative model for the dislocation generation hypothesis, preliminary calculations were made of the local stress concentrations developed at second-phase particles (and voids) in an isotropic matrix subjected to external hydrostatic pressure.

In addition to the direct observation of dislocation generation in iron, a preliminary study was made of commercial purity, powder metallurgy tungsten - as an important refractory metal which is brittle in the annealed condition at room temperature and pressure - to determine its response to pressure cycling. The results showed that after pressure cycling annealed tungsten wire to 13 kilobars, the ductile-brittle transition temperature (determined from constant strain-rate

tensile studies at temperatures from 27^o - 200^oC) exhibited a decrease of some 50^oC compared with that for similar uncycled material. Corresponding to these observations, the stress-strain curves of the pressure cycled wires showed a decrease in yield-drop, although the drop was not completely eliminated. The structure of the fine wires (0.030 in. diameter) could not then be examined by transmission electron microscopy, but the study of thin foils prepared from tungsten sheet made from the same type of stock showed that the concentration of impurity particles was extremely low, which was considered to be a possible factor in the failure to eliminate the yield-drop. Similar purity sheet from another manufacturer showed a much higher concentration of such particles. Analogous marked variations in particle content were observed in thin foils prepared from commercial-purity molybdenum sheet obtained from powder metallurgy and from arc-melting stock.

During the second year of the program (beginning 1 June 1965), the objectives have been to (a) study pressure effects on iron and iron-carbon alloys as being a model system for the investigation of the mechanism of dislocation generation under hydrostatic pressure and of the variables controlling the magnitude and nature of the effect; and (b) continue the study of the effect of pressure on ductile-brittle behavior by investigating the influence of sub structure and second-phase particles in tungsten and tungsten-thoria alloys. This extended investigation has involved a second graduate assistant, Mr. P. Trester who joined the research program in September 1965, in addition to the first graduate assistant, Mr. G. Das, who joined the program in September 1964. In addition, H. Ll. D. Pugh, the Head of the Plasticity Division, National Engineering Laboratory, U.K., who is particularly well known for his studies of mechanical behavior and forming processes at high pressure,

has been appointed as the Republic Steel Corporation Distinguished Visiting Professor at Case for the academic year 1965-66. During this year he is participating in the current study of pressure effects on structure and mechanical behavior of bcc metals.

The present report describes the research carried out during the six-month period beginning 1 June 1965. During this period, the principal research effort has been directed to the effects of pressure cycling up to 25 kilobars on ductile-brittle transition behavior in tungsten and tungsten- 1% thoria alloys. In addition, measurements of pressure effects on the discontinuous yield phenomena in high purity iron-carbon alloys have been initiated as part of the study of quantitative relationships between pressure, dislocation structure and changes in the yield characteristics.

2. TUNGSTEN AND TUNGSTEN-THORIA ALLOYS

Subsequent to the previously reported⁽⁷⁾ observation of a decrease in the ductile-brittle transition temperature (T_d) of annealed tungsten wire specimens (Material W-3, 0.030 in. diameter) brought about a pressure cycle to 13 kilobars, a more detailed study of the transition behavior of this material was undertaken with the objective of determining the magnitude of the effect more precisely. These earlier measurements were made on specimens annealed in vacuum in a tantalum-strip resistance furnace (Brew Model No. 1064) at temperatures of 1310° and 1650°C, and subsequently reduced by electropolishing to 0.025 in. diameter along a 1 inch gage length. The tensile tests, carried out in an Instron machine at a crosshead speed of 0.025 in. min.⁻¹ from room temperature to 200°C, showed that T_d was greater than 200°C for both the annealing temperatures used. In the present study, a larger number

of specimens (56) was annealed as a single batch to minimise possible variations in properties due to any slight differences in conditions in successive annealing runs. The preparation, measuring and testing of tensile specimens from these was carried out in the manner described previously⁽⁷⁾, except for the use of a rotary electropolishing jig designed to allow up to 22 specimens to be electro-machined simultaneously. As well as speeding up the preparation process, this technique facilitates the control of the finished gage diameter to the desired constant value.

The results of tensile tests on three such batches annealed at 1310°C are shown in Figure 1 in the form of yield stress, fracture stress and reduction in area versus test temperature. As can be seen, the results for the different batches plot on the same curves. However, in contrast with the previous data for this annealing temperature, the transition temperature is much lower - in the region of 80°C rather than above 200°C - and the yield stress at a given test temperature is also lower. Furthermore, tensile tests on specimens from these three batches which were cycled to pressures of 14 and 25 kilobars after annealing, showed no change in yield or transition behavior, within the experimental error, from the as-annealed specimens. The hydrostatic pressure cycling experiments were conducted in a modified piston-cylinder apparatus in the manner described previously^(6,7). To ensure identical test conditions, successive tensile tests at a given temperature were carried out alternately on "as-annealed" and "annealed and pressure-cycled" specimens; whenever possible, a minimum of two tests for each condition was used. The results for similar tests carried out on specimens annealed at 1600°C and pressure cycled to 15 kilobars, shown in Figure 2, also gave a lower transition temperature (190°C) than that found previously for a similar annealing temperature of 1649°C ($T_d > 200^\circ\text{C}$).

Likewise, no change in yield or transition behavior was apparent for the annealed and pressure-cycled specimens.

The difference in yield and transition behavior from the earlier results for nominally similar annealing temperatures suggested that the actual temperatures were higher in the earlier annealing experiments. Metallographic examination of longitudinal micro-sections along the diameter of wires from the various annealing batches has confirmed this view. Figure 3a shows the long ribbon-shaped grains or "fibers" observed in the specimens annealed at 1310°C (corresponding to the tensile data given in Figure 1) and Figure 3b the coarser-grained, more equi-axed structure of the specimens annealed at 1600°C (Figure 2 data). The former temperature is in the range in which light and transmission electron microscopy observations⁽⁹⁾ have recently established that annealing occurs by strain-induced migration of the boundaries of the long ribbon-shaped grains (fibers) present in the as-drawn wire. In the region of 1500°C, this leads to the formation of a more equiaxed grain structure and at higher temperatures growth and coarsening of this structure takes place. Thus, the structures found in the present specimens correspond closely to these observations. In contrast, the earlier annealed structures show anomalously coarse grains for the temperatures intended (Figure 3c - nominally 1310°C, and Figure 3d - nominally 1649°C). Comparison of the microstructures and properties indicates that the actual temperatures at which these structures developed were both well above 1600°C. It is now believed that the incorrect temperatures resulted primarily from erroneous thermocouple readings due to contamination of the couple used at the time of these experiments.

The different responses to pressure-cycling shown by the different annealed structures suggests that the pressure-induced lowering of the

transition temperature may be quite specific in that it will occur in this commercial purity tungsten only when recrystallisation and grain growth has proceeded to the point at which the structure has a low concentration of dislocation-sources which can become operative on straining the material. That is, the material is source-poor, as is generally the case for annealed iron. (The possibility that slight variations in impurity existed between the earlier and later wires tested cannot be discounted, but must be considered unlikely). The variable response of tungsten to pressure cycling encountered here is not unique among results reported for pressure effects on mechanical behavior. Thus, Bullen et al (10,11) have indicated that the 'ductilising' of chromium by pressure cycling can also be specific to particular batches of the metal. Attempts to reproduce the effect in at least three other laboratories are known to have been unsuccessful. Bullen et al also observed that the ductility of pressure-cycled chromium is sensitive to the strain rate used in the tensile testing - annealed chromium cycled to 10 kilobars was found to be ductile when tested at 0.002 min^{-1} , either ductile or brittle at 0.005 min^{-1} , and brittle at 0.01 and 0.05 min^{-1} . However, this does not appear to be as important a factor in the case of the tungsten discussed above, since reducing the strain rate (crosshead travel) by a factor of 10 from the normally used rate of $0.025 \text{ in. min}^{-1}$ to $0.0025 \text{ in. min}^{-1}$ gave no measurable change in the ductility or other tensile characteristics (see Figure 2 for the tungsten annealed at 1600°C).

The nature of the yield drop in tungsten is of particular importance to the general problem of the factors controlling flow and stress and yield phenomena in this metal. The appearance of distinct yield drops in polycrystalline tungsten is well known to be erratic (see for example ref. 12) and it has not yet been clearly associated with a

particular interstitial impurity element. In high purity single crystals, as prepared by electron-beam zone-refining techniques, discontinuous yield is observed only for crystals oriented with their axis of stressing close to $\langle 110 \rangle$ orientation - other orientations show smooth homogeneous yield at considerably lower stress levels⁽¹³⁾. In this case, the yield characteristics do not appear to be associated with the presence of impurity elements and the controlling mechanism is as yet unknown. A possible factor complicating the interpretation and understanding of yield in polycrystalline tungsten of commercial purity is that annealing textures in tungsten frequently, and particularly for wires, exhibit $\langle 110 \rangle$ directions parallel to the direction of deformation.

Analogous studies of the effects of pressure cycling on the tensile behavior of tungsten containing a deliberately added second phase, a tungsten - 1% thoria alloy (Material W-4; commercial purity 0.030 in. diameter wires prepared by powder metallurgy techniques) have been initiated for various annealing conditions. Specimen preparation and testing is being carried out in a similar manner to that used for the tungsten, except for the use of higher annealing temperatures and appropriate modifications to the electro-polishing method for shaping the tensile specimens. The results of tensile tests up to 225°C on tungsten - 1% thoria wires vacuum annealed at 2000°C and 2200°C are given in Figures 4 and 5 respectively. It is seen that the transition temperature (T_d) increases from some 130°C for the 2000°C annealed condition to 180°C for the 2200°C condition. Pressure cycling specimens to 15 and 20 kilobars respectively for these two conditions gave no measurable change in the magnitude of the transition temperatures. Light micrographs of longitudinal cross-sections from specimens annealed at these two temperatures (Figure 6) show that the 1% thoria alloy annealed at 2000°C (Figure 6a) still exhibits a "fiber" structure of only slightly

wider elongated grains than shown in tungsten annealed at 1310°C. Annealing at 2200°C results in a coarser elongated grain structure (Figures 6b and 6c), but with more indications of fiber structure remaining than observed in tungsten annealed at the much lower temperature of 1600°C (Figure 3b). Thus, the optical microstructures indicate that even at 2200°C recrystallisation of the alloy is not yet complete so that, as discussed earlier for some of the tungsten, the pressure-induced dislocation generation mechanism may be inoperative or ineffective. As for the tungsten, clarification of this point should come with the study of material annealed at higher temperatures and the examination of the dislocation structure of the wires by thin foil electron microscopy.

3. IRON-CARBON ALLOYS

Following the previously reported thin foil electron microscopy observations of pressure-induced generation of dislocations in iron-carbon alloys in the spheroidised^(6,8) and quench-aged⁽⁷⁾ conditions, the study of this model system has been continued with measurement of the corresponding effects on discontinuous yielding in tension. This portion of the research is being undertaken principally by Mr. Trester, who joined the program in September 1965.

The earlier measurements of terminal yield behavior were carried out on machined tensile specimens of small size (0.05 in. gage diameter by 0.33 in. gage length)⁽¹⁾ or on small diameter wires (0.048 in. diameter)⁽²⁾: In order to improve the accuracy of the stress-strain measurements in the microstrain and yield regions and to facilitate thin-foil microscopy observations directly on the tensile specimens, a larger specimen size was selected for the current work. The tensile specimen is a 1-3/4 in. long button-head type, with a 0.150 in. gage

diameter and a 1 in. gage length. This large size will permit, in particular for specimens stressed below the macro-yield region, a correlation of sub-structural changes with alterations in initial plastic behavior induced by pressure cycling. It is anticipated that this approach will clarify the dynamic role of the fresh dislocations and/or sources introduced.

The initial tensile measurements have been directed to establishing suitable specimen preparation and testing procedures, and to measuring the suppression of yielding in these high purity iron carbon alloys compared with that in the commercial plain carbon steels investigated previously⁽¹⁾. Currently, the specimens are machined from stress-relieved bar (590°C for 1 hour), vacuum annealed at 667°C ± 5 for 1 hour and finish ground. Before tensile testing, all specimens are checked for dimensions and alignment on a INDI - AC electronic gage indicator. Two types of tensile loading rigs in an Instron constant strain-rate machine were tried on the annealed specimens - a simple extension bar device fitted directly to the fixed and moving stages of the machine, and a concentric axial loading rig designed to facilitate the self-alignment of the specimen. In the stress-strain observations with these rigs, strain was measured both from cross-head travel and from electrical resistance strain gages, and concentricity of loading was determined from 4 strain gages placed at 90° intervals around the specimen diameter. The results showed that the axial loading rig gave considerably more consistent results, especially for the upper yield behavior, which is particularly sensitive to misalignment. Consequently, despite the greater complexity and difficulty of using this device, it was adopted for subsequent tests of pressure cycling effects. It is well known that even when care is taken over the axially of loading, the upper yield point is

a difficult parameter to measure accurately and reproducibly. However, for the present purposes, quantitative comparisons of different conditions can most usefully be made from measurements of the lower yield or a flow stress, which can be measured accurately.

The effects of pressure cycling on the yield behavior of an annealed iron - 0.065 wt.% carbon alloy tested in the axial rig are illustrated in Figure 7 for maximum pressures of 8.7 and 20 kilobars. The successive lowering of the stress for yielding and the elimination of the upper yield point at the highest pressure are apparent (these curves represent the average of two tests each for 8.7 and 20 kilobars and 5 tests for the as-annealed material. A further feature of the curves, particularly marked for the highest pressure, is the apparent deviation from a linear stress-strain relationship at comparatively low stresses. This result is qualitatively in keeping with the dislocation - generation hypothesis, but more precise strain measurements in the low stress region are needed for a quantitative analysis. Such measurements are to be made. The changes in yield stress from that at atmospheric pressure are shown in Figure 8 as a function of peak pressure. The results for the 0.065%C alloy are in keeping with those expected although the present data is too limited to define the pressure dependence of the decrease in yield stress - in particular the existence of an initial critical pressure and a saturation effect at higher pressures. The nature of the results confirms the choice of the high purity iron-carbon alloys as a model material. In contrast, an annealed 0-18% carbon steel (AISI 1018) showed very little change in yield characteristics after subjection to a pressure of 15 kilobars (see Figure 8). This unexpectedly small response is being investigated further for its possible relation to the substructure developed by the particular annealing process used for this steel.

4. FUTURE WORK

During the next six-month period of the program, effort will be continued on the effects of pressure on the ductile-brittle transition behavior of commercial purity tungsten and tungsten-thoria alloys. Particular attention will be paid to the nature of the dislocation substructure of the annealed material as defined by thin foil electron microscopy, changes induced in it by pressure, and the relation of these to transition behavior. Some work on annealed rod specimens will be conducted in an attempt to clarify the possible influence of preferred orientation. In the case of the iron-carbon alloys, the study of the changes in yield behavior as a function of volume proportion of the second phase and peak pressure will be continued. In the alloy showing the maximum effects, attempts will be made to relate quantitatively the nature of the dislocation structure and the yield behavior, especially in the microyield range.

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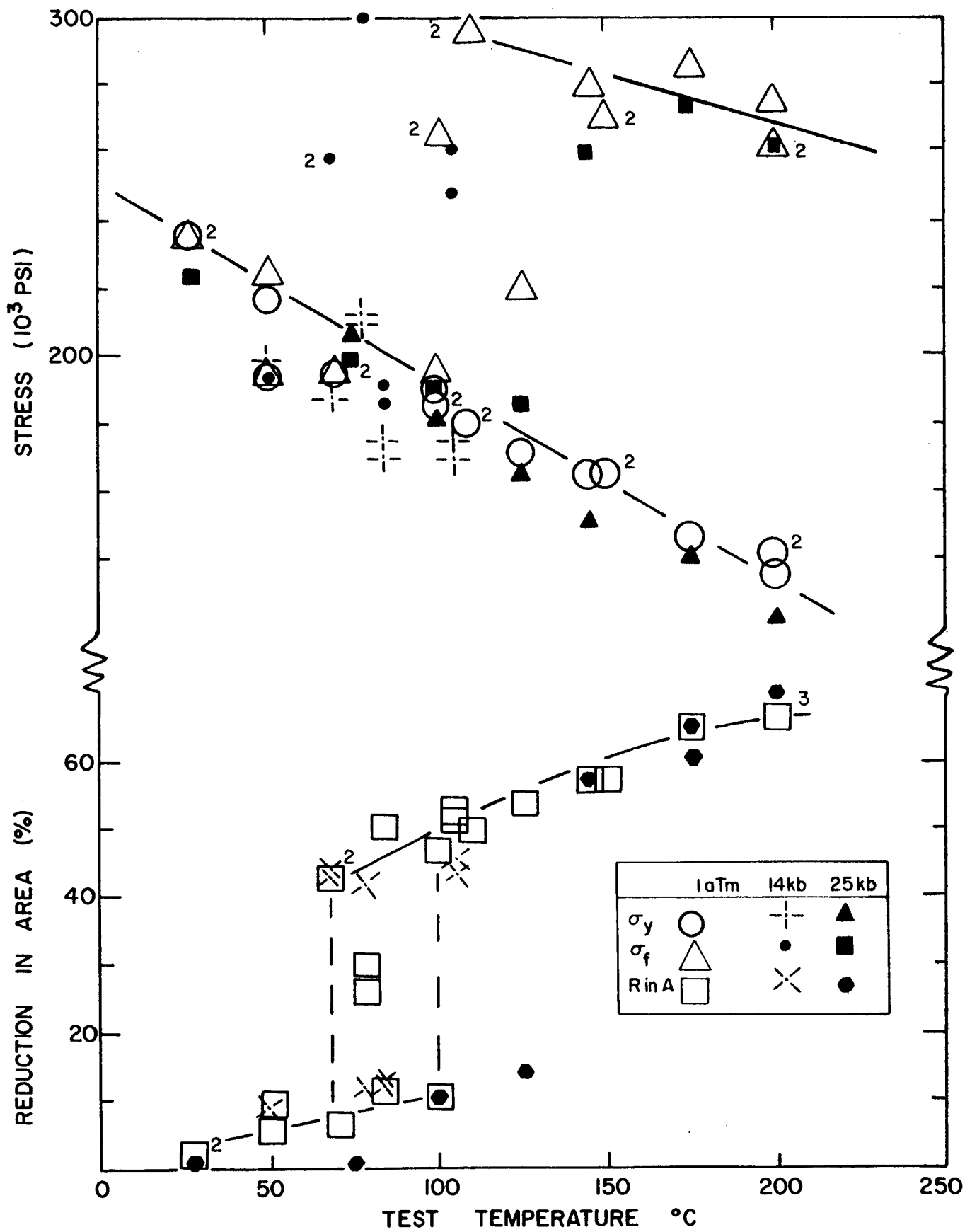


FIG. 1: Effect of test temperature on tensile properties of tungsten wire annealed at 1310°C. (a) As-annealed, (b) cycled to 14 kilobars, (c) cycled to 25 kilobars.

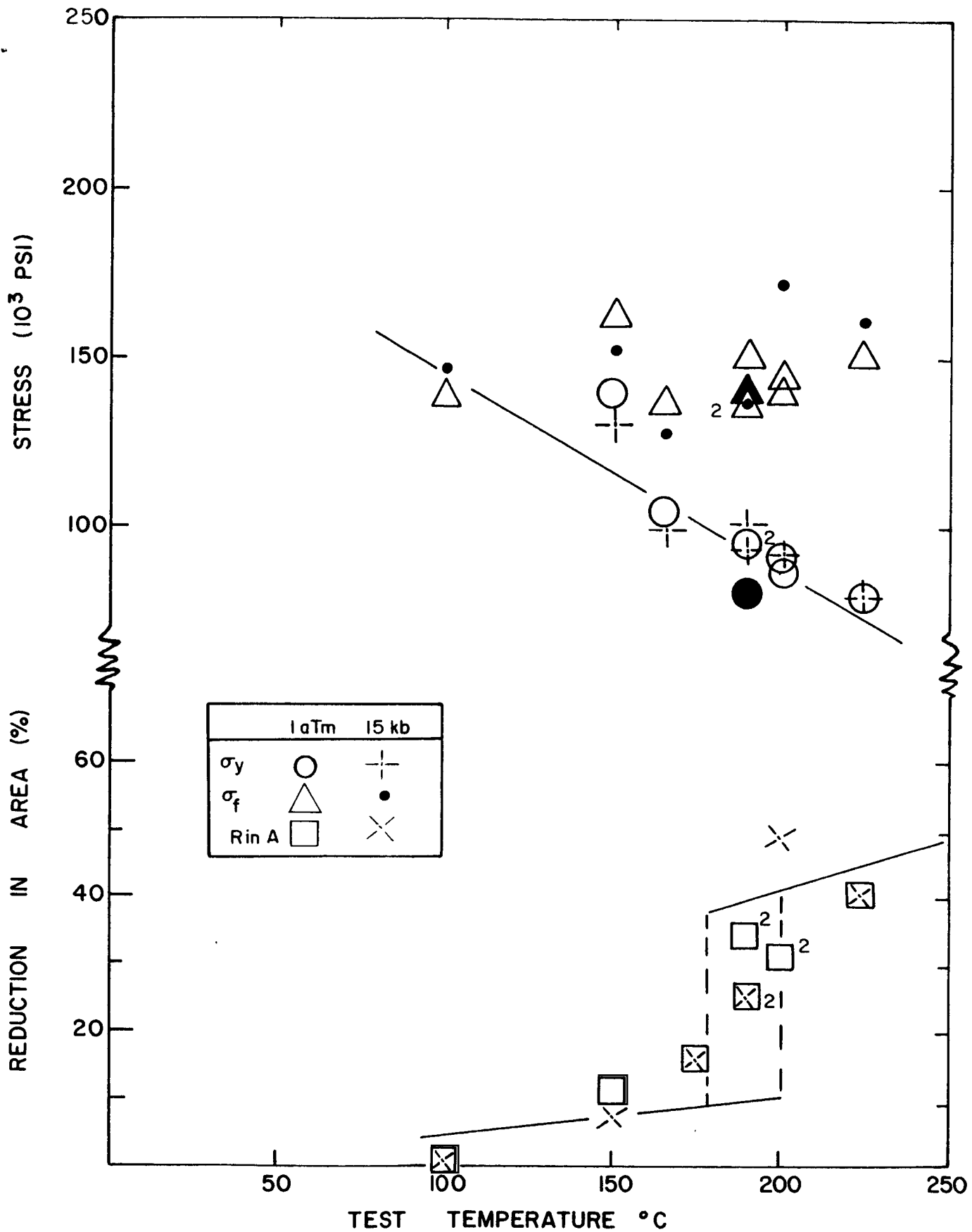
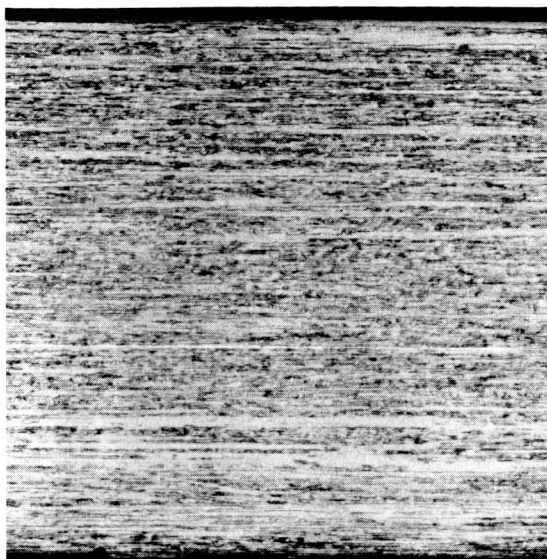
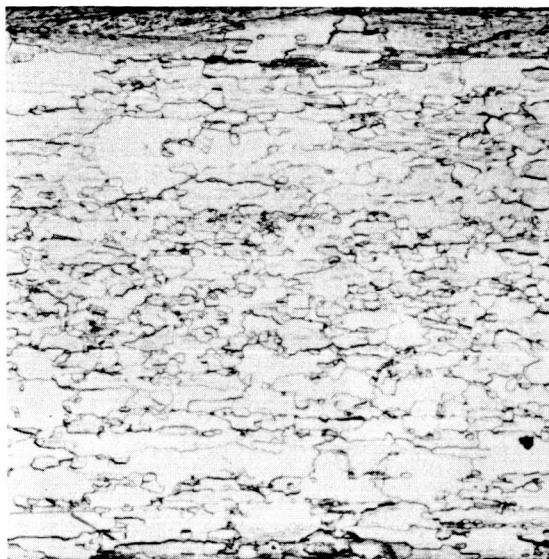


FIG. 2: Effect of test temperature on tensile properties of tungsten wire annealed at 1600°C. (a) as-annealed, (b) cycled to 15 kilobars. Solid data points refer to crosshead travel at 0.0025 in. per min. All other data at 0.025 in. per min.



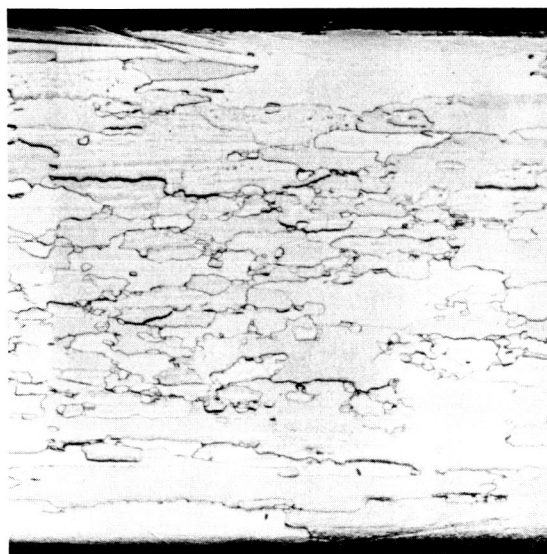
100X

(a) Annealed at 1310°C.



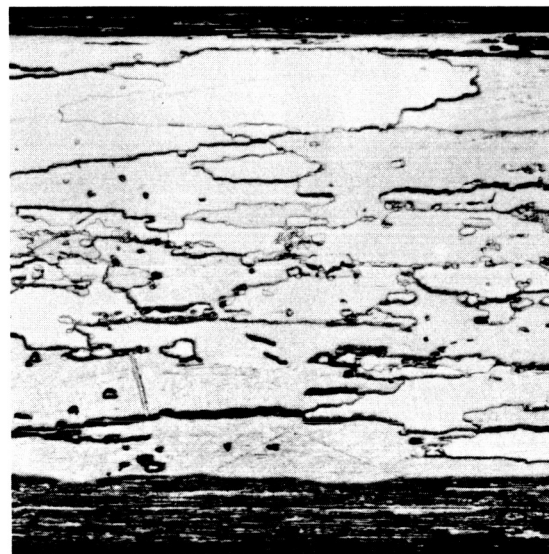
100X

(b) Annealed at 1600°C



100X

(c) Nominally annealed at "1310°C".



100X

(d) Nominally annealed at "1649°C".

FIG. 3. Microstructures of tungsten wires annealed at indicated temperatures. Longitudinal sections.

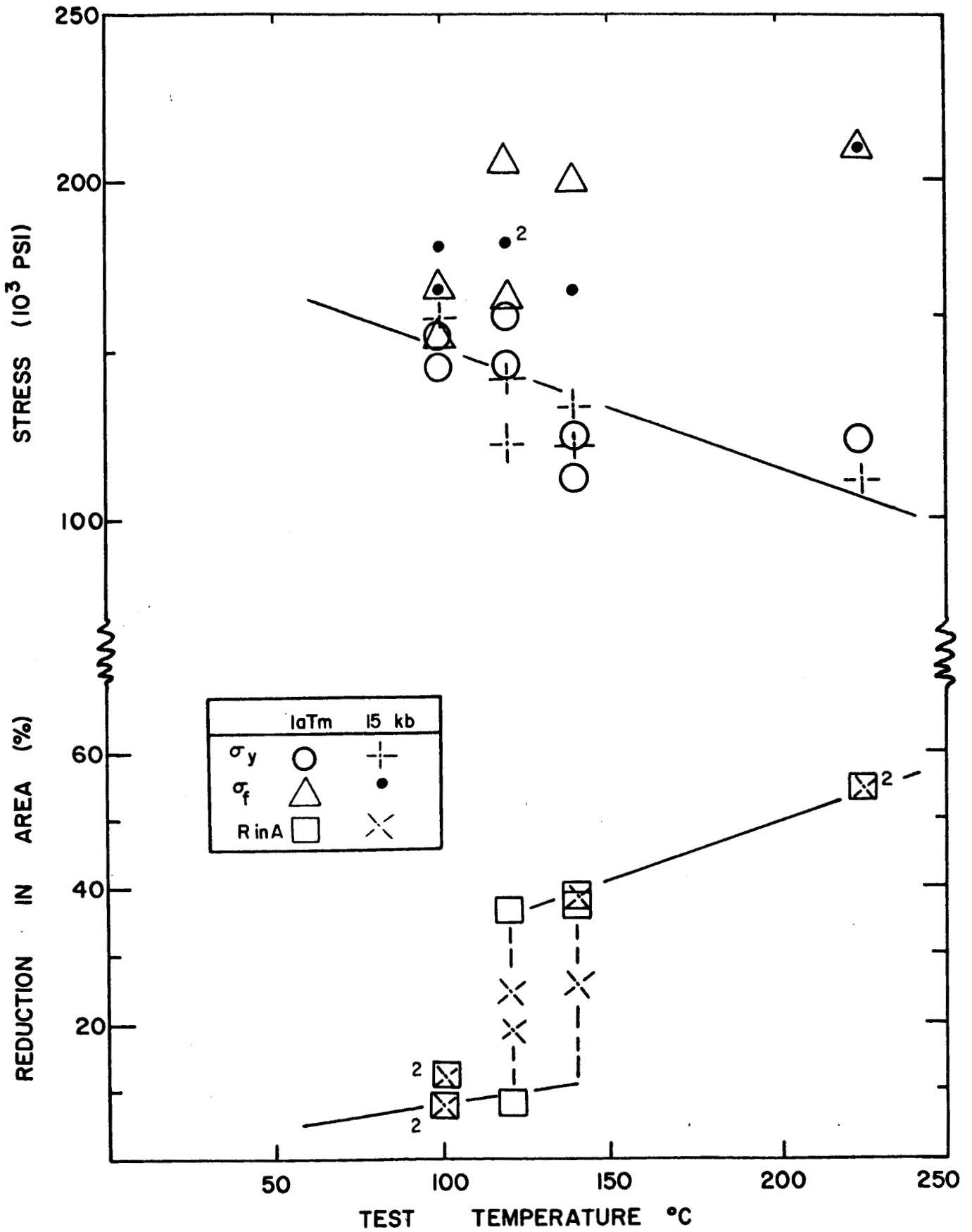


FIG. 4: Effect of test temperature on tensile properties of tungsten-1% thoria wire annealed at 2000°C.
 (a) as-annealed, (b) cycled to 15 kilobars.

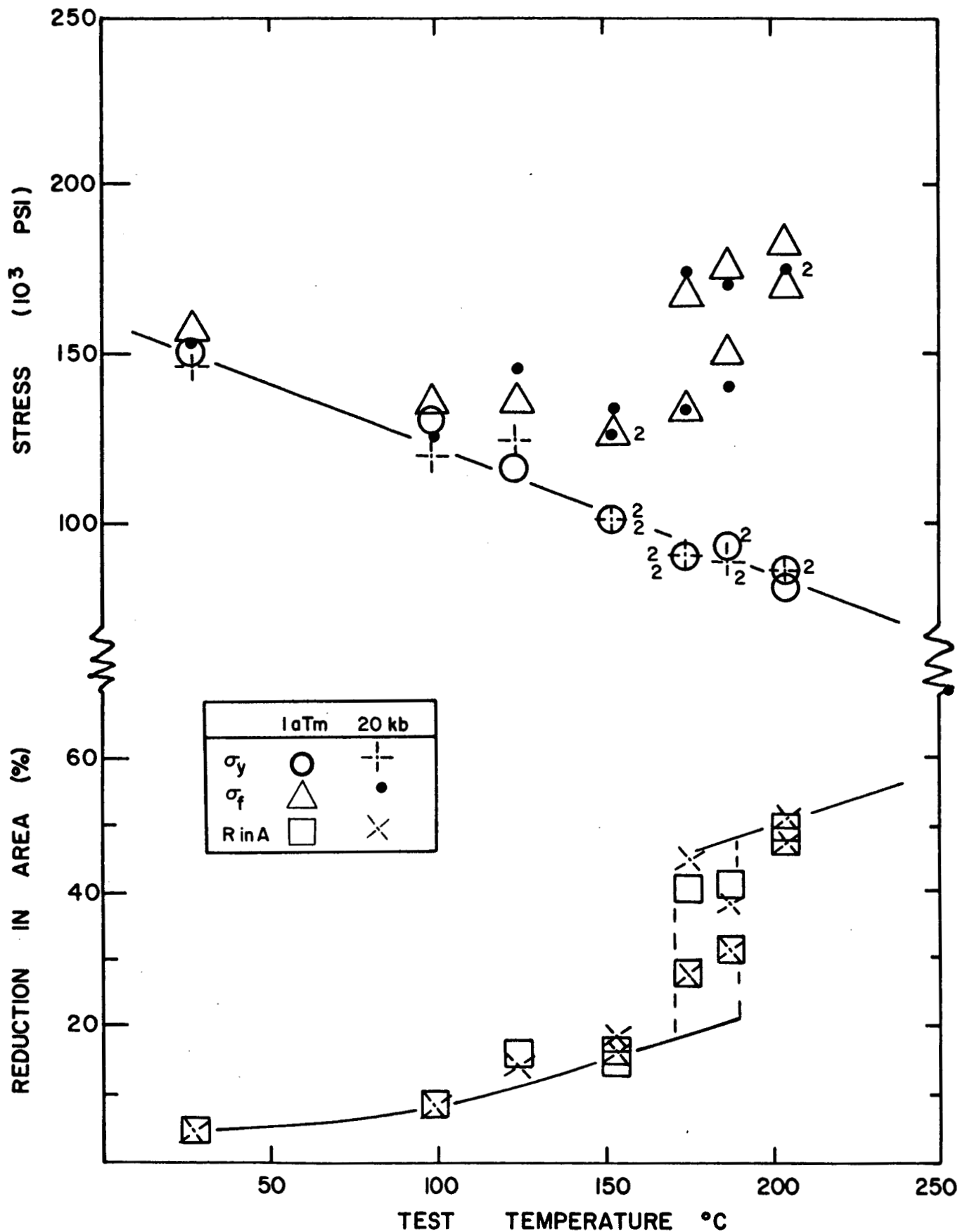
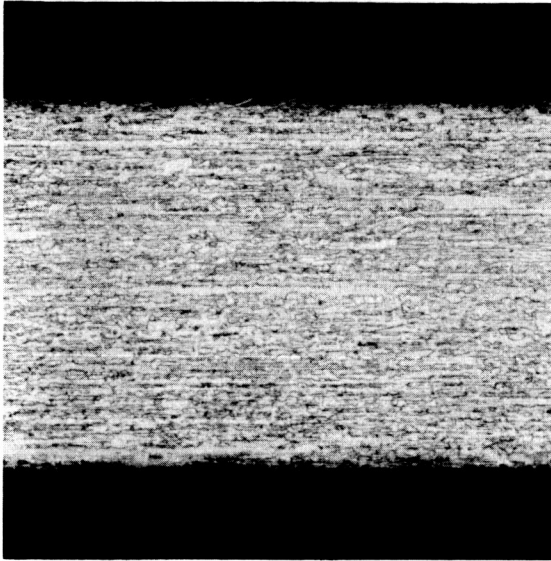
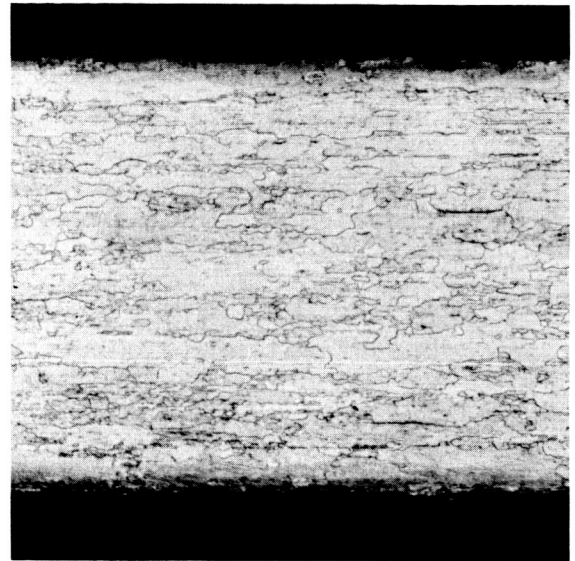


FIG. 5: Effect of test temperature on tensile properties of tungsten-1% thoria wire annealed at 2200°C. (a) as-annealed, (b) cycled to 20 kilobars.



X100

(a) Annealed at 2000°C.



X100

(b) Annealed at 2200°C.



X500

(c) Annealed at 2200°C

FIG. 6. Microstructures of tungsten - 1% thoria wires annealed at indicated temperatures. Longitudinal sections.

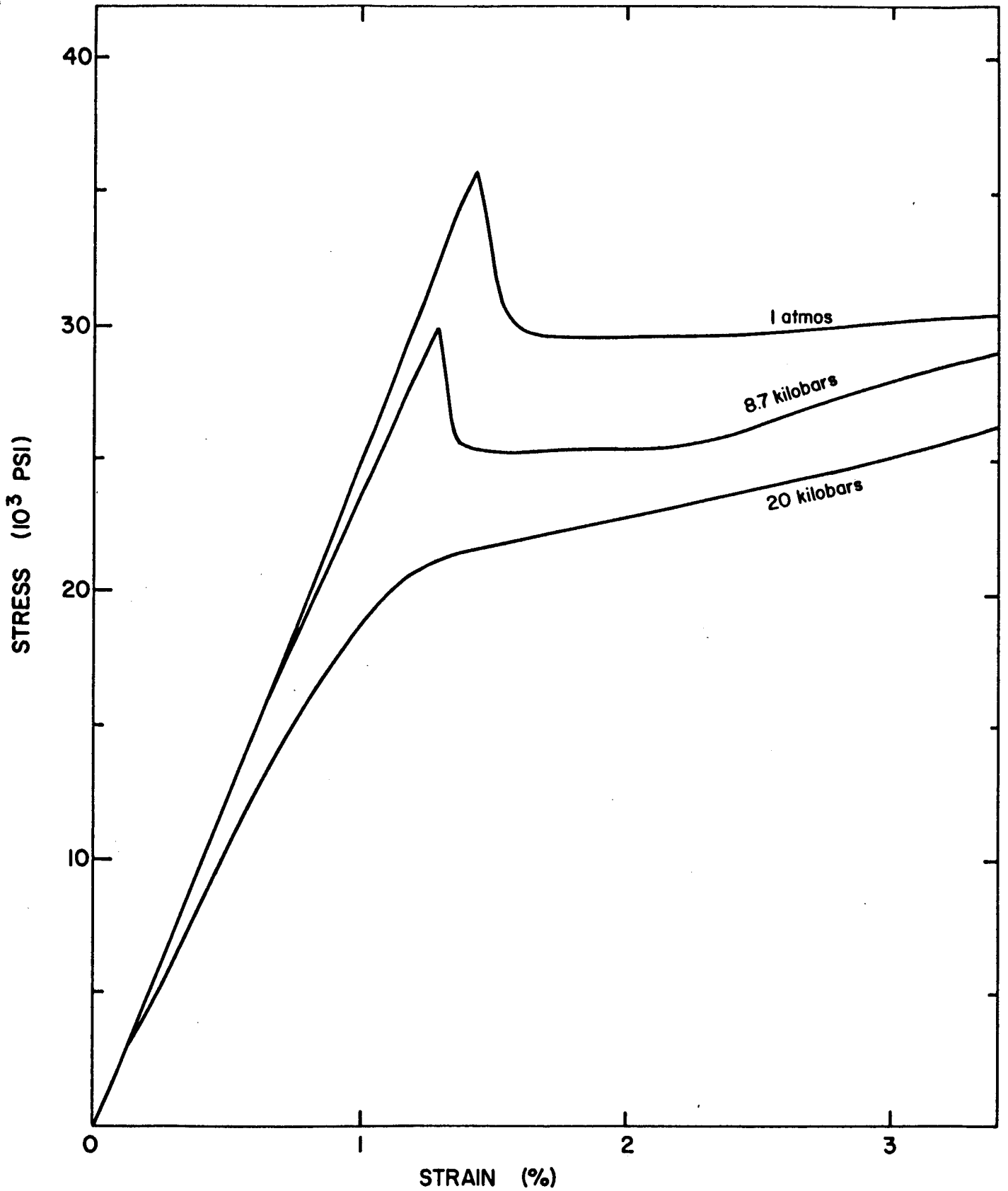


FIG. 7: Effect of cycling to indicated pressures on tensile yield behavior in annealed iron - 0.065 wt.% carbon alloy (averaged curves - see text).

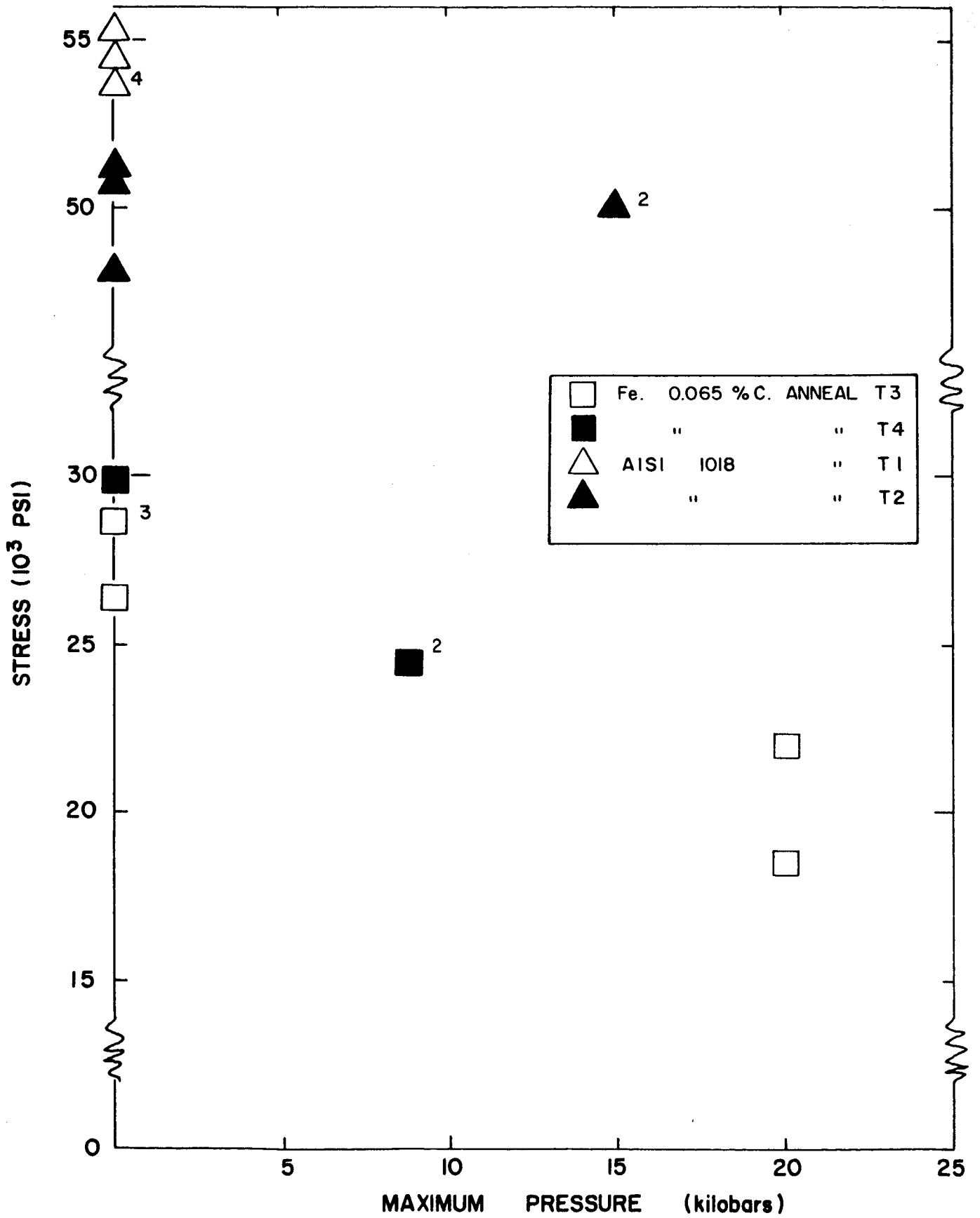


FIG. 8: Dependence of yield stress on maximum pressure in the cycle.