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TITLE: MECHANICAL PROPERTIES OF 800-MeV PROTON-IRRADIATED METALS

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MECHANICAL PROPERTIES OF 800-MeV PROTON-IRRADIATED METALS[2]

ABSTRACT: Accelerator beam line components and spallation neutron targets operate in an irradiation environment where changes in mechanical properties can adversely affect component integrity. The present work presents a preliminary study of the effects of low fluences (10^{19} to 10^{20} p/cm^2) of 800-MeV protons on the yield strength, tensile strength, and ductility of samples of 304 stainless steel, Alloy 718, molybdenum, and tantalum. Tensile samples (0.75 or 1.6 mm thick) were directly water cooled during irradiation and were tested at room temperature. For the 304 stainless steel and annealed Alloy 718, the yield strengths increased by about a factor of 3 and 1.6, respectively, while the ductility decreased ~30 and 40 percent. In the bcc metals (tantalum and molybdenum) the yield strengths increased by at least a factor of 2. Tantalum samples retained significant ductility at room temperature, while several molybdenum specimens broke at less than 0.2 percent strain. These irradiation-induced changes at low proton fluences should not impair the usefulness of these materials (other than molybdenum) in accelerator environments.

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KEY WORDS: accelerator, spallation neutron source, proton irradiation, tension test, strain hardening parameters, stainless steels - 304, tantalum, molybdenum, Inconel 718.

High current particle accelerators such as the Los Alamos Meson Physics Facility (LAMPF) can produce significant radiation damage in beam line components that either intercept the proton beam directly or receive a high flux of scattered particles. At LAMPF, components such as the beam line window that separates the vacuum of the main beam line from air receive the direct proton flux, which, after a number of years operation, can reach a fluence of 10^{23} p/cm^2. Irradiation by the direct proton beam also generates significant internal heating in such components, requiring that they be water cooled. The beam line window, for example, consists of two 0.2-m-diam plates, separated by a 1-mm-thick gap that allows continuous circulation of cooling water. Water pressures near 2.1 MPa (300 psi) impose additional stresses and require that the window remain a sound pressure vessel despite changes in mechanical properties resulting from the irradiation. By careful design, it has generally been possible to limit temperature rises in such components to about 300 K maximum. A second component that is directly irradiated is the beam stop, consisting of copper plates inside a 304 stainless steel cylinder 0.2 m diam and 0.6 m long. As with the window, gaps between the plates allow water circulation to remove the heat deposited by the beam. Integrity of the stainless cylinder is important in preventing large water leaks that could result in loss of cooling and melting.

In addition to the components considered above, interest has been expressed in using high atomic mass targets irradiated by the proton beam as spallation neutron sources [1]. These targets, using such materials as tungsten and depleted uranium, cannot be directly water cooled, because radiolysis of the water can result in high corrosion rates. Since these components and targets must maintain structural integrity in this hostile environment of stress, temperature, and irradiation, there is a need to characterize the property changes under these conditions.

In this paper, we examine mechanical property changes in 304 stainless steel and Alloy 718, both useful as structural materials for beam line components, at low proton fluences. Type 304 stainless steel is the material used for the beam stop cylinder and faces, while Alloy 718 was chosen as a higher strength material that could be welded and was used in several models of the beam line window. Tantalum was of interest as a spallation neutron source target while being directly water cooled, since it is not strongly corroded by the products of cooling-water radiolysis. Molybdenum, although strongly attacked by radiolysis products, was chosen to allow comparison of mechanical property changes with tantalum.

EXPERIMENTAL PROCEDURE

The approach taken in studying changes in mechanical properties was to machine sheet tensile samples (see Fig. 1) from the materials of interest and stack the corrosion-resistant samples on an open fixture (Fig. 2), which was then placed in a water-cooled sample holder and proton irradiated. Following irradiation, the samples were removed from the fixture in a hot cell, gamma scanned, and tensile tested.

Irradiation

For irradiation, samples (secured in their fixtures) were placed in water-cooled target holders used by the isotope production facility. The target holders are at the end of 9-m-long horizontal, shielded penetrations known as stringers, and are inserted just upstream of the beam stop. Target holders are attached to the stringers remotely, and the stringers have no provision for electrical feedthroughs; consequently, sample temperatures could not be directly monitored. The high water flow rate in direct contact with the samples, together with knowledge of volumetric heating produced by the proton beam, allowed us to calculate the maximum temperature rise for the various materials. For this calculation, the beam current was assumed to be 500 μA and a heat transfer coefficient of 0.5 $W/cm^2 \cdot °C$ between the sample surface and the flowing water was assumed. The maximum temperature at the midsection of each metal tested is given in Table 1, assuming the cooling water to be at 293 K. Accelerator operation is interlocked with water flow in the isotope production stringers, so that a sharp drop in flow immediately shuts down the proton beam.

To monitor the proton fluence received by the tensile samples, aluminum foils were placed in line with the tensile samples. Following irradiation, the atomic fraction of ^{22}Na produced by the 800-MeV protons was determined by gamma spectroscopy and the proton fluence was calculated assuming a production cross section of 1.2×10^{-2} barn. Both the aluminum foils and the actual samples were also counted using a scanning gamma spectrometer in which a narrow slit is stepped along the sample while the total number of counts (independent of energy) is recorded. This provides a measure of the variation in proton fluence from one end of a sample to the other, as well as the variation from one sample to another. A typical variation in fluence along the gauge length for a stainless steel sample was 20 percent. For samples stacked one behind the other in the proton beam, fluences varied by at most 9 percent, but samples in different positions in the holding fixture received fluences differing by factors of 2. Determination of the absolute fluences is expected to be within ±50 percent.

Tensile Testing

The samples were highly activated following irradiation (often several hundred R/h), requiring all handling and testing to be done remotely in a hot cell. Following disassembly of the specimen fixture and the gamma scanning, the spacing of a previously placed pair of scribe marks was checked prior to tensile testing. Samples were then placed in the grips and the loading pins inserted. An extensometer was clipped onto the gauge section and the sample pulled to 1 percent strain, at which point the test automatically halted. The extensometer was removed and the test was resumed at constant crosshead speed until the load peaked and started to drop. The load was then removed from the sample and the spacing between scribe marks was measured and used to determine the uniform elongation. Strain values were derived from the load-time curve by setting the strain at maximum load equal to the uniform strain measured using the scribe marks.

RESULTS

Table 1 presents the irradiation conditions of proton fluence, displacements per atom (dpa), and maximum sample temperature. The dpa values were computed by using the damage energy cross sections computed by Coulter et al. [2]. Changes in yield strength (at 0.2 percent offset), tensile strength, and uniform strain are shown for all metals tested in Table 2. In general, irradiation to the fluences attained here produces a large increase in the metal's yield strength and a smaller increase in tensile strength. It was not possible to define the tensile strength for molybdenum because only one of four samples elongated beyond 0.01 strain without fracturing brittlely. In all cases, irradiation reduced the uniform strain, with the greatest decreases occurring in the bcc metals. Figure 3 compares engineering stress-strain curves for unirradiated and irradiated 304 stainless steel. Following irradiation, a yield drop is present in the stress-strain curve. Changes in the stress-strain curves for molybdenum and tantalum following irradiation are shown in Fig. 4. These bcc metals show yield drops prior to irradiation, and exhibit low uniform strain following irradiation, particularly molybdenum.

For the 304 stainless steel, the engineering stress-strain data were converted to true stress-true strain data. These data were then fitted to the power law strain hardening equation $\sigma = K\epsilon^n$, where K is the strength coefficient and n is the strain hardening exponent. Curves of log (true stress)-log (true strain) were concave in shape so that the values of n and K were found by a least-square fit to the data between true strains of 0.05 and 0.5. Values of the strength coefficient, strain hardening exponent, and uniform strain, ϵ_u, are given in Table 3 for unirradiated and irradiated 304 stainless steel.

DISCUSSION

In general, yield strengths are increased more than tensile strengths, resulting in a reduction in strain hardening. This is quite noticeable from the stress-strain curves for 304 stainless steel, and is quantitatively shown by the decrease in strain hardening exponent. This exponent should be numerically equal to the uniform strain if the stress-strain curve fits the equation $\sigma = K\epsilon^n$. Comparing preirradiation values of n and ϵ_u shows good agreement, but following irradiation n is 19 percent less than ϵ_u, indicating that the true stress-true strain curve is concave, with a higher slope at higher strains [3]. We did not choose to quantitatively compare strain hardening behavior between yield and 0.05 true strain, because data in this range were obtained assuming the strain to be proportional to the crosshead displacement, which is not a good assumption at strains less than 0.05. A qualitative examination of the true stress-true strain curves (at strains between 0.002 and 0.05) shows the strain hardening exponent to be significantly decreased in the irradiated samples. A factor of 2 increase in fluence led to a small (7 percent)--but significant--increase in yield strength. Work is now underway to prepare transmission electron microscopy specimens of irradiated 304 stainless steel in both the undeformed and strained states. Examination of the undeformed material should clarify the nature of the damage, while the strained material will be examined for the presence of features such as dislocation channeling.

A qualitative comparison can be made between our results on changes in yield strength, tensile strength, and uniform elongation, and the results of others [4] for neutron-irradiated samples. In that work, 304 stainless steel samples were neutron irradiated at temperatures less than 373 K to neutron fluences between 1.2×10^{13} and 1.4×10^{20} n/cm^2 (E > 1 MeV). Examination of those stress-strain curves shows a yield drop at fluences of 1.3×10^{19} n/cm^2 and above, similar to the drop we observe. At a fluence of 2×10^{19} n/cm^2, the yield strength increased by a factor of 2.2, the tensile strength by 1.1, and the uniform strain by 0.7. For our samples, these factors were 2.9, 1.2, and 0.75 - suggesting that the same proton fluence may have been slightly more effective in strengthening the material. A more valid comparison would require knowledge of the neutron fluence as converted to displacements per atom. Comparison of the stress-strain curves for this study with others suggests that the yield stress is raised by irradiation-produced vacancy clusters and dislocation loops. The presence of such defect agglomerates has been shown to produce similar features in the stress-strain curves of fcc and bcc materials [5,6].

For the Alloy 718 samples, irradiation of the annealed material raised the yield stress, while irradiation of hardened material lowered it. For the annealed material, the increase in yield strength is undoubtedly caused by dislocation interaction with the irradiation-produced microstructure. The decrease in yield strength and tensile strength following irradiation of the precipitation-hardened material is not presently understood. Previous results [7] at fluences of 5-10 x 10^{21} n/cm^2 and an irradiation temperature of 673 K, with testing at room temperature, have shown either little change or an increase in the yield and tensile strengths. Our unirradiated heat-treated material showed uncharacteristically low elongation, which may be due to an improper heat treatment.

In addition to large increases in yield strength, irradiation of the bcc metals tantalum and molybdenum reduced the uniform strain for both metals. For tantalum, all samples were still capable of about 14 percent strain following irradiation to 1.5 x 10^{20} p/cm^2 (1.5 dpa). Strain hardening was greatly reduced compared with the unirradiated material, and the yield strength was doubled. For molybdenum samples tested after irradiation to

1.5×10^{19} p/cm^2 (0.08 dpa) the yield strengths had more than doubled, but only one of four samples reached 0.02 strain before fracturing. Other workers [8] have observed similar decreases in strain following irradiation and room-temperature testing. It is likely that the ductile-brittle transition temperature has been raised above room temperature by the irradiation.

SUMMARY

Samples of 304 stainless steel irradiated to low proton fluences near ambient temperature display a 200 percent increase in the yield strength and a 50 percent decrease in uniform strain. These changes are similar in magnitude to those observed during neutron irradiations at similar temperatures, and will not impair the usefulness of this material for beam line components. A similar conclusion applies to the annealed Alloy 718, which showed a 60 percent increase in yield strength and a 40 percent decrease in uniform elongation. A testing program is required to extend these results to higher fluences and to temperatures in the 600 K range.

The use of tantalum for spallation neutron targets is possible from a materials standpoint, although the ductility will likely decline further at higher fluences. Molybdenum is clearly a poor choice for beam line components, because it becomes brittle at room temperature following even a short-term irradiation.

TABLE 1—Irradiation Conditions
Maximum Temperatures and Proton Fluence

Metal	Sample Thickness (mm)	Maximum Temperature (K)	Proton Fluence (p/cm^2)	Fluence (dpa)
304 Stainless Steel	1.6	335	2×10^{19}	0.06
Alloy 718	1.6	340	2.2×10^{19}	0.[illegible]
Tantalum	0.7	330	1.5×10^{20}	1.50
Molybdenum	0.8	320	1.3×10^{19}	0.08

TABLE 2 — Preirradiation and Postirradiation Mechanical Properties

	Yield Strength				Tensile Strength				Uniform Plastic Strain (%)	
	Preirradiation		Postirradiation		Preirradiation		Postirradiation		Pre-	Post-
Material	MPa	(psi)	MPa	(psi)	MPa	(psi)	MPa	(psi)	Irradiation	Irradiation
304 Stainless Steel Annealed[a]	134	(19 500)	383	(55 000)	516	(74 900)	602	(87 300)	77	58
Alloy 718 Annealed[b]	552	(80 000)	889	(129 000)	1 017	(147 500)	1 040	(150 800)	38	23
Alloy 718 Heat Treated[c]	1 570	(228 000)	1 410	(205 000)	1 700	(247 000)	1 520	(220 000)	2	2
Tantalum Stress Relieved[d]	141	(20 500)	290	(42 000)	230	(33 300)	285	(41 350)	36	14
Molybdenum Stress Relieved[d]	290	(42 000)	690	(100 000)	383	(55 500)	-	-	19	2

[a] 0.5 h at 1310 K, rapidly gas cooled.

[b] 0.5 h at 1225 K, gas cooled.

[c] 0.5 h at 1225 K, aged 8 h at 990 K, furnace cooled to 894 K and held for total aging time of 18 h.

[d] 0.5 h at 1373 K, slow cooled.

TABLE 3 — Strain Hardening in 304 Stainless Steel

Condition	Strength Coefficient K MPa (psi)		Strain Hardening Exponent, n	Uniform Strain, ε_u
Unirradiated Annealed	1 170	(169 700)	0.53	0.57
Irradiated	1 200	(174 000)	0.35	0.46

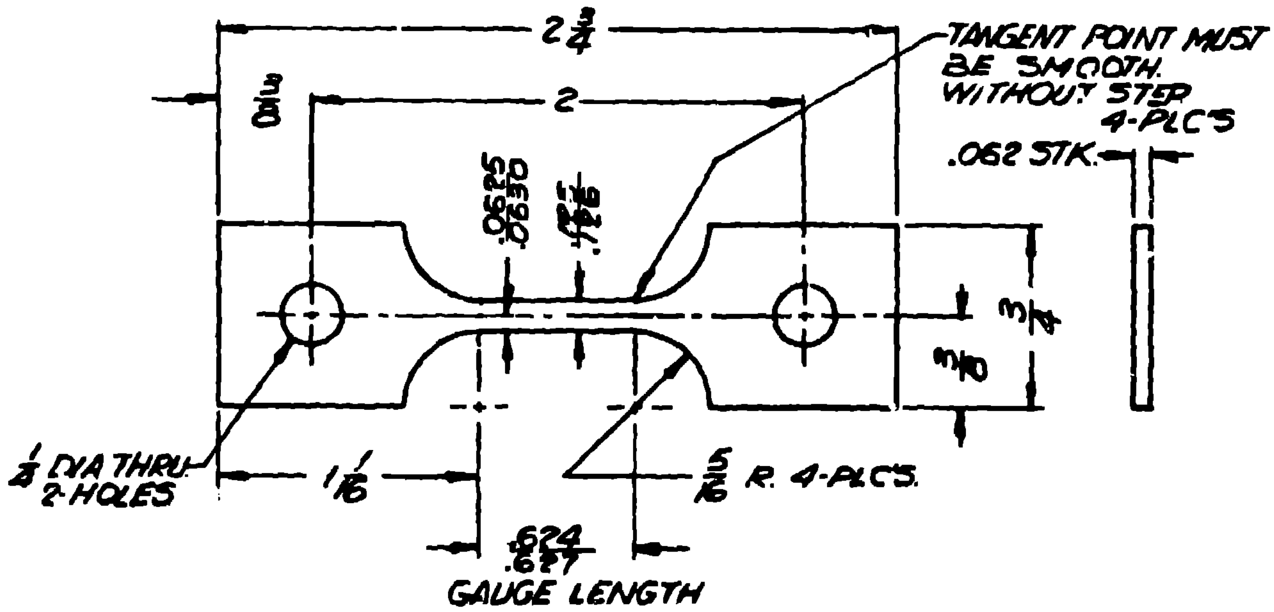

FIG. 1—Dimensions of a typical tensile sample. Nonstandard specifications were required to fit the samples into the water-cooled target holder for proton irradiation.

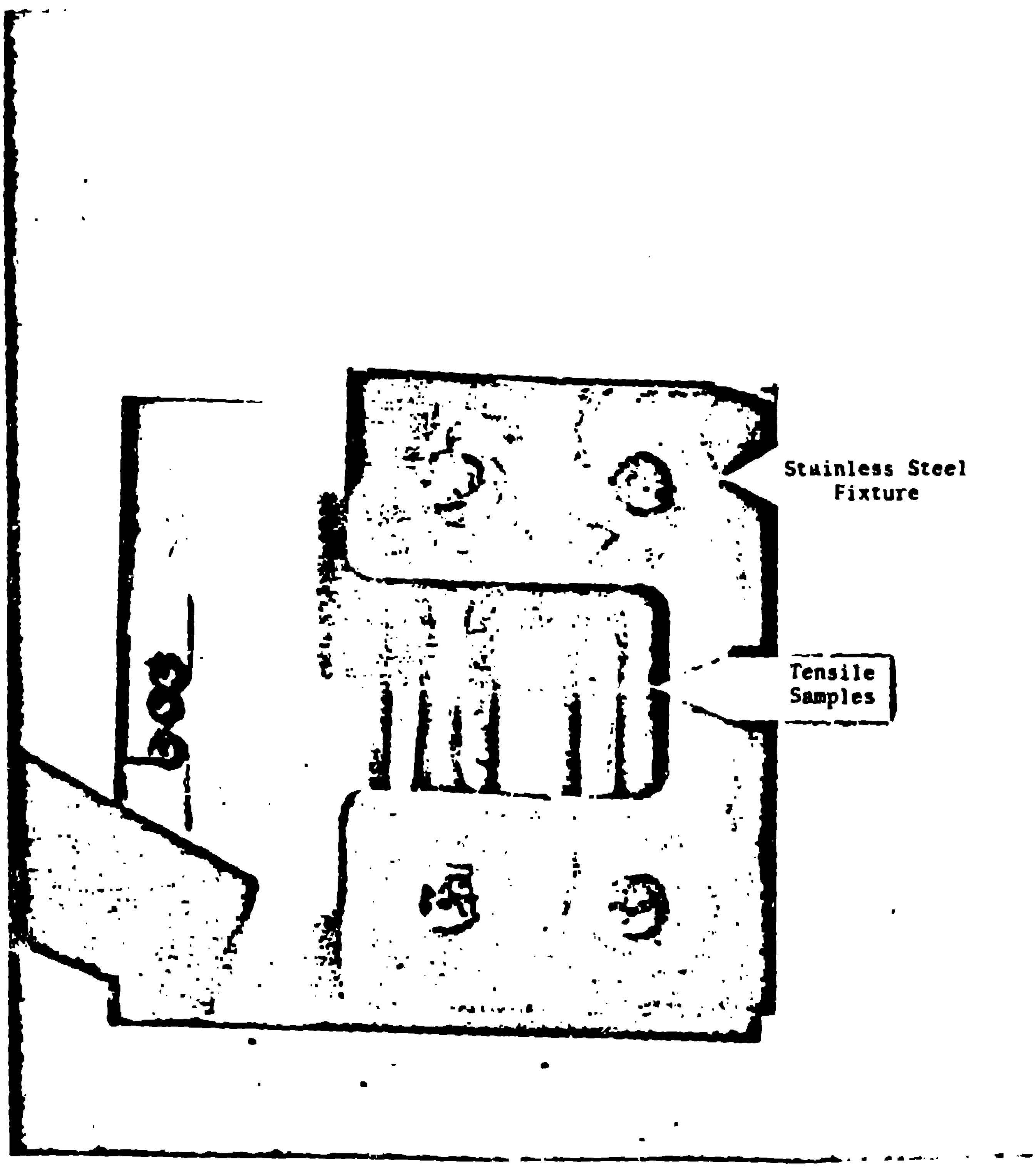

FIG. 2 — S' stainless steel fixture holding tensile samples following irradiation. Spacers separate the tensile samples to ensure adequate water flow during irradiation.

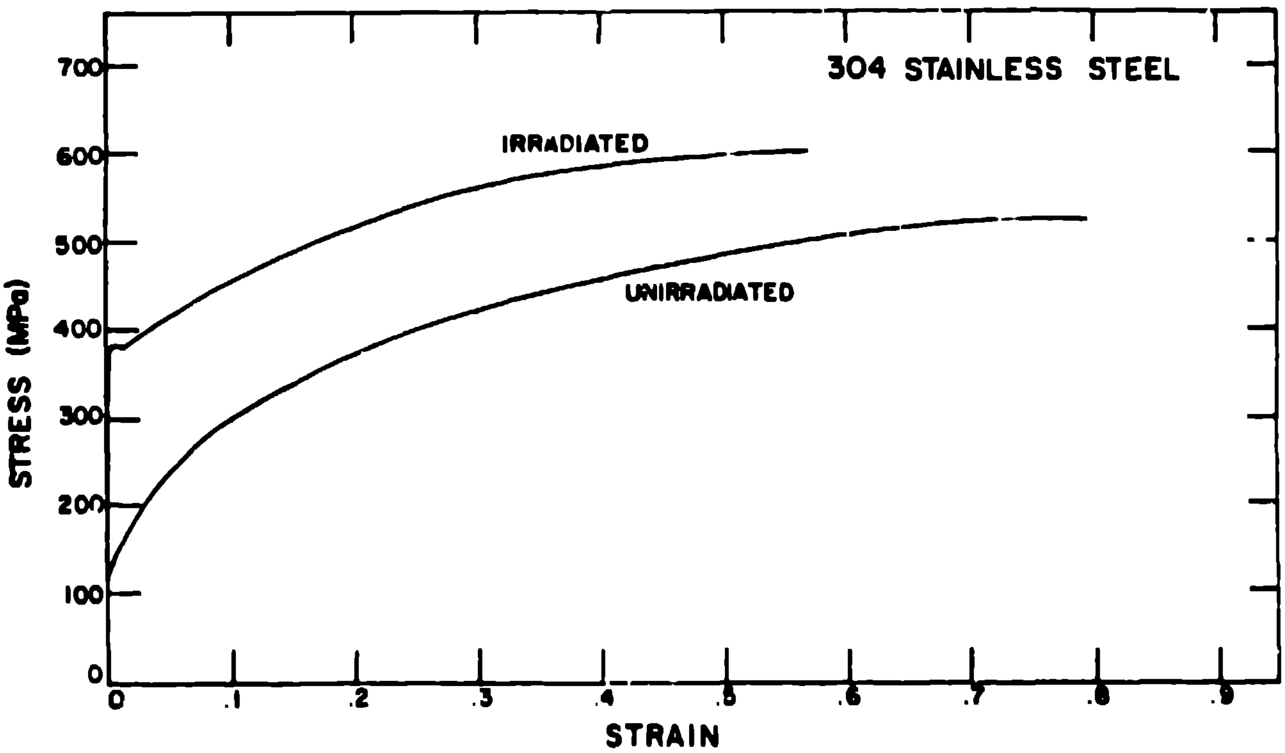

FIG. 5 — Typical engineering stress-strain curves for 304 stainless steel, both unirradiated and irradiated to a fluence of 2×10^{19} p/cm^2.

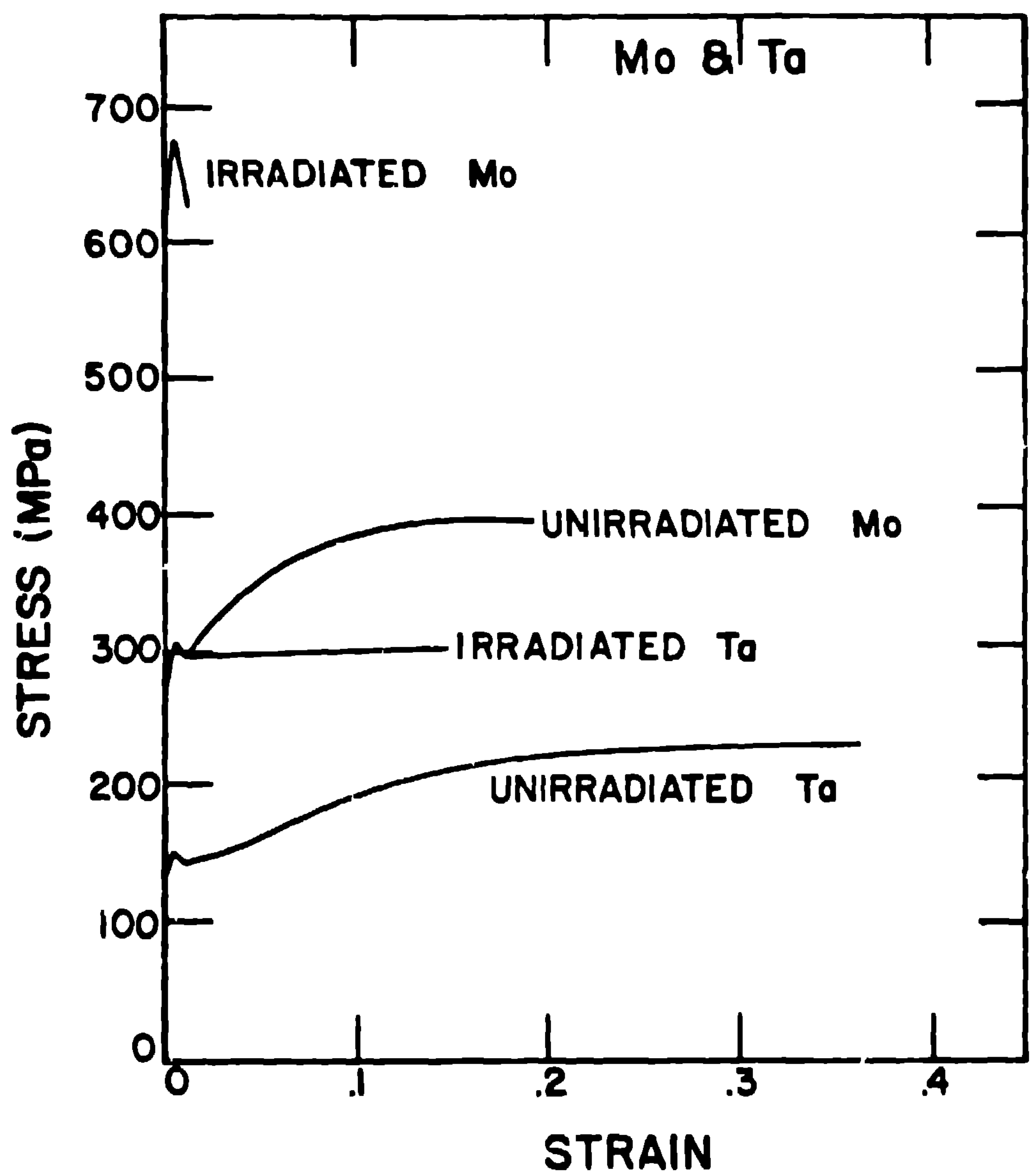

FIG. 4 — Typical engineering stress-strain curves for molybdenum and tantalum, both unirradiated and irradiated to fluences of 1.3×10^{19} and 1.5×10^{20} p/cm^2, respectively.

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