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SUMMARY

Grain-refining techniques using multistep heat treatments to reduce the grain size of five commercial high-strength steels were investigated. The goal of this investigation was to improve the low-temperature toughness as measured by Charpy V-notch impact test without a significant loss in tensile strength. The grain size of four of five alloys investigated was successfully reduced to 1/10 of original size or smaller with increases in Charpy impact energy of 50 to 180 percent at -320°F. Tensile properties were reduced from 0 to 25 percent for the various alloys tested. An unexpected but highly beneficial side effect from grain refining was improved machinability.

INTRODUCTION

The development of two high-Reynolds-number cryogenic wind tunnels (Langley 0.3-Meter Transonic Cryogenic Tunnel (TCT) and the National Transonic Facility (NTF)) has imposed stringent requirements on materials for wind-tunnel model construction. Working stresses on high-performance aircraft models for the NTF can approach 100 ksi with temperatures near -320°F. The toughness-versus-strength trend for commercial alloys (fig. 1 taken from ref. 1) shows the difficulty of material selection due to the divergent requirements of high strength and high toughness.

Since many commercial high-strength steels need additional fracture toughness to meet current criteria for cryogenic models, Langley Research Center conducted a program to improve the cryogenic toughness of commercial high-strength steels through grain-refining heat treatments (refs. 2 and 3). The relationship between fine grain size and improved low-temperature toughness has been long established; however a suitable technique for developing this fine-grained structure in high-strength martensitic and maraging steels was desired.

J. W. Morris and co-workers at the University of California at Berkeley developed grain-refining heat treatments for the ferritic alloys Fe-12Ni and 9Ni (refs. 4 and 5) and demonstrated an excellent combination of strength and toughness for these alloys at cryogenic temperatures. The goal of the Langley program was to realize similar improvements in the higher strength martensitic alloys, particularly HP 9-4-20 and 18Ni 200 grade maraging steel.

EXPERIMENTAL PROCEDURE

The program was divided into four study phases as follows:

1. An attempt to duplicate Morris's data for the ferritic alloy 9Ni

2. Development of grain-refining techniques for the high-strength quenched and tempered martensitic alloys HP 9-4-20 and HP 9-4-30 manufactured by Republic Steel Corp.

3. Extension of the technique to the pseudo-maraging steel AF-1410

4. Further extension to the fully maraging steel 18Ni 200 grade (VascoMax C-200, manufactured by Teledyne Vasco)

Measure of Material Toughness

Because of the embrittling effect of cryogenic wind-tunnel operations, Hudson (ref. 6) proposed that a minimum fracture toughness of K_{1c} of 85 ksi-in$^{1/2}$ be required for cryogenic model and support hardware at test temperature. Since Charpy impact data are more readily available than fracture toughness data, Hudson further proposed that the Barson-Rolfe relationship be used to screen candidate materials:

$$K_{1c} = \left[2E(C_{vn})^{3/2}\right]^{1/2}$$

where E is Young's modulus (psi) and C_{vn} is the Charpy impact energy (ft-lb). This relationship, while questionable for high-toughness austenitic steels, appears to give good correlation for the ferritic and martensitic steels. For example, Newman and Lisagor's (ref. 7) measurements of K_{1c} for 18Ni 200 grade steel agreed closely with the value predicted by the Barson-Rolfe equation of 63 ksi-in$^{1/2}$ at -320°F for C_{vn} = 17 ft-lb (a typical value observed for plate stock of this alloy).

Since the Barson-Rolfe equation gives C_{vn} = 25 ft-lb for K_{1c} = 85 ksi-in$^{1/2}$, Langley "Wind-Tunnel Model Systems Criteria" (LHB 1710.15) specifies 25 ft-lb as the minimum impact energy for high-strength (greater than 95 ksi) cryogenic structural materials at test temperature. Thus, the principal goal of the grain-refining program was to meet the toughness criterion of C_{vn} = 25 ft-lb while maintaining acceptable tensile strength.

Test Specimens

All materials used in this investigation were production alloy plate stock. Specimen sizes for all alloys except AF-1410 were 12 × 12 in. with thicknesses of 3/4 to 1 1/8 in. Because of the limited amount of material available, the AF-1410 experiment was conducted on standard 10-mm Charpy specimens which had been cut from a 5/8-in-thick cross-rolled plate. The specimen sizes and condition prior to grain refining are listed in table I. The chemical composition of each alloy is shown in table II.

Grain Refinement Procedure

Morris et al. (ref. 5) reported large reductions in the grain size of 9Ni steel by subjecting the material to multiple heating and cooling cycles, alternating between the austenitic (γ) and the dual-phase austenite plus ferrite ($\gamma + \alpha$) region followed by rapid cooling (water quench). This procedure was used in this investigation, with the grain-refining temperatures being determined from the alloy phase diagram when available and from the iron-nickel equilibrium and transformation diagrams (figs. 2 and 3, taken from ref. 8).

Test Procedure

The following tests were used to evaluate the effectiveness of the grain refinement procedure.

Microstructure.- Metallographic specimens were prepared for each alloy to compare the stock (control) grain structure with the grain-refined structure. Photomicrographs were made at magnifications of × 100 and × 400 for each alloy to show the reduction in grain size. Although all others could be successfully etched with oxalic acid, 5 percent nital, and aqua regia, the two alloys showing the largest grain size reduction (HP 9-4-20 and 18Ni 200 grade) proved to be very difficult to etch in the grain-refined condition. HP 9-4-20 could be etched by nital after 15 to 20 minutes, but 18Ni 200 grade showed no attack by nital after 60 minutes. A suitable etchant was acid ferric chloride (10g ferric chloride, 100 ml hydrochloric acid, 100 ml distilled water). All alloys investigated were more difficult to etch in the grain-refined condition than in the stock condition.

Charpy impact tests.- Standard (10 × 10 × 55 mm) Charpy V-notch specimens were prepared according to ASTM A 370 (ref. 9) from both stock and grain-refined material for all alloys. Tests at -320°F were conducted by soaking specimens for 1 hour in liquid nitrogen (LN_2) and breaking them immediately upon removal. Most breaks occurred within 5 sec after removal, and data from breaks occurring more than 7 sec after removal were disallowed. Both the absorbed energy and the lateral expansion were recorded.

Tensile tests.- Tensile tests were performed on HP 9-4-20 and 18Ni 200 grade in both the stock and the grain-refined condition. The specimen geometry is shown in figure 4. This specimen is a modified version of the ASTM A 370 (ref. 9) subsize tensile specimen with pin-jointed ends to allow it to be tested while submerged in LN_2. Tests at -320°F were performed by soaking the specimens in LN_2 for 1 hour and testing them with a modified Instron tensile test machine using an insulated LN_2 container and pin and clevis connection in place of the normal specimen grips. All tests were conducted at a crosshead speed of 0.020 in/min. Yield strength was determined by the 0.2-percent offset method from the load displacement curve. Ultimate strength and reduction in area were also recorded. The test setup precluded measurement of actual elongation of the gage length; however total crosshead travel was recorded. Though it is not a true measure of elongation, the crosshead travel is indicative of relative changes in elongation between the stock and grain-refined materials.

Hardness.- Rockwell hardness tests were conducted on all materials in both the grain-refined and the stock condition. The hardness values for the stock and grain-refined material were compared to estimate relative changes in tensile strength for those materials on which tensile tests were not performed.

Fracture mode.- Scanning electron microscope fractographs were made of the Charpy fracture surfaces on HP 9-4-20 and 18Ni 200 grade to determine the fracture mode at -320°F.

EXPERIMENTAL RESULTS

9Ni Steel (ASTM A353-79)

The as-received 9Ni steel had a fine-grained martensitic structure with an average grain size of 20-25 μm. The micrographs in figure 5 are typical. One of the specimens was subjected to the heat treatment used by Morris et al. (ref. 5). The heating and cooling schedule for this treatment is shown in figure 6 and is designated GR I. The resultant microstructure (fig. 7) was very fine grained, similar in appearance to the stock material but with the grains reduced in size to 4-7 μm. The hardness of this specimen was 23 R_c compared to 21 R_c for the control specimen.

The second specimen was subjected to the same grain-refining schedule as GR I except the tempering time (step 6) was increased from 1 hour to 4 hours. This procedure (GR II) resulted in larger grains than those of the GR I specimen. The grain size of the GR II specimen was only slightly smaller than the grain size of the control specimen, and its hardness decreased to 20 R_c.

The third specimen received the same grain-refining schedule as GR I except the tempering temperature (step 6) was lowered from 1050°F to 900°F. This heat treatment (GR III) resulted in very fine grain size (5-10 μm) similar to the grain size of the GR I specimen, but its hardness was only 18-19 R_c.

Thus the best combination of grain refinement and strength retention was obtained with the GR I specimen. Charpy impact specimens were then cut from the GR I and the control specimen for testing at both room temperature and -320°F. During the machining of the Charpy specimens, both materials appeared tough, but readily machinable. The grain-refined specimens (GR I) machined somewhat easier than the control specimens; with the GR I material, higher removal rates and smoother surface finishes were obtained during milling operations.

The Charpy impact energy at -320°F averaged 73 ft-lb for the control specimens and 102 ft-lb for the GR I specimens (an increase of 40 percent). Room temperature tests showed an increase of 12 percent for the GR I material. At -320°F, lateral expansion was 58 mils for the GR I material and 45 mils for the control material. Measurements of the GR I lateral expansion were complicated by the extreme deformation of the fracture surfaces and the large number of incomplete fractures (4 out of 8) (see fig. 8). Test data for the GR I and control specimens are summarized in table III.

These results are consistent with Morris's findings (ref. 5) that the cryogenic toughness of 9Ni steel can be improved through multistep heat treatment to refine the grain size.

Martensitic Alloys HP 9-4-20 and HP 9-4-30

Experiments were conducted to determine whether the techniques used for ferritic 9Ni could be applied to quenched and tempered martensitic alloys with higher carbon content. The martensitic alloys HP 9-4-20 and HP 9-4-30 were chosen for these experiments. Langley has successfully used both of these alloys for conventional (noncryogenic) wind-tunnel model systems where medium to high (125-175 ksi) tensile strengths are required, but because of low toughness, neither material has been considered a viable candidate for cryogenic service. HP 9-4-20, while retaining some degree of toughness at cryogenic temperatures, fails to meet the toughness criterion of $K_{1c} \geqslant 85$ ksi-in$^{1/2}$ (see fig. 1) and is inferior to 18Ni 200 grade in both strength and toughness. Also, both HP alloys have been characterized by Langley fabrication personnel as very difficult to machine to close tolerances.

HP 9-4-20.- The nickel content of this alloy was 9.15 percent compared with 8.90 percent for 9Ni steel (see table II), so similar transformation temperatures could be expected. The phase transformation diagram (fig. 9 taken from ref. 10) showed phase change equilibrium temperatures of 1185°F (A_{e1}) and 1375°F (A_{e3}) for material of similar composition. Figure 10 shows the GR I heat treatment schedule for 9Ni with the A_{e1} and A_{e3} temperatures superimposed. The austenitization temperature (steps 2 and 4) and the two-phase decomposition temperature (steps 3 and 5) fall near the upper and lower limits of the dual-phase region. This would indicate that the grain refinement cycles (steps 2-5) should be raised approximately 75°F. Because the effect on grain refinement of the additional alloying elements in HP 9-4-20 was unknown, the first attempt was made using the GR I treatment without alteration.

The as-received material had a tempered martensitic structure with an average grain size of 80-90 μm (ASTM size 4). The grains, while relatively uniform in size and shape, displayed a distinct laminar subgrain structure. The micrographs in figure 11 are typical of the as-received material.

The first specimen was subjected to the GR I treatment shown in figure 10. The resultant microstructure revealed a significant reduction in grain size. The structure appeared to be very homogeneous with ultrafine grains of approximately 5 μm. Very little structural detail could be resolved even at magnification as high as × 800. The hardness of this specimen was 32-33 R_c compared with 39 R_c for the control specimen. This specimen was designated GR 20-I. The micrographs in figure 12 are typical of the fine grain structure produced by the GR I heat treatment.

The second specimen (GR 20-II) was subjected to the GR II grain-refining schedule, with the tempering time (step 6) being 4 hours. The microstructure produced by this treatment showed a large variation in grain size (20-80 μm) and lack of the homogeneity observed in the GR 20-I specimen. This observation is consistent with the results for 9Ni; long tempering times tend to erase the grain-refining process. Typical structure produced by the GR 20-II heat treatment is shown in figure 13.

Charpy impact and tensile test specimens were fabricated from the GR 20-I and the control specimen for testing at both room and cryogenic temperatures. The control specimen exhibited the poor machining properties typical of HP alloys, but the GR 20-I specimen did not. The machinability of the grain-refined material was significantly improved. Langley fabrication personnel judged its machinability equivalent to that of AISI 400 series stainless steels.

The Charpy impact energy at -320°F was 38-39 ft-lb with lateral expansion of 20 mils for the GR 20-I material and 13-14 ft-lb with lateral expansion of 6 mils for the control material. The GR 20-I specimens exhibited a pronounced shear lip and a fully ductile fracture surface (see fig. 14).

The tensile test at -320°F recorded an average yield strength of 160.5 ksi with an ultimate strength of 224.6 ksi for the GR 20-I material. The average elongation was 0.345 in. with a reduction in area of 40.8 percent. The GR 20-I specimens showed considerable plastic yielding prior to fracture at -320°F (see fig. 15). Test data for the GR 20-I and control specimens are summarized in table IV. No further experiments were performed on HP 9-4-20 in this investigation.

HP 9-4-30.- The degree of grain refinement and the large increase in Charpy impact energy obtained with HP 9-4-20 was somewhat unexpected because of its higher carbon content (0.19 percent compared with 0.07 percent for 9Ni). Morris et al. (ref. 11) have reported that the rapid quenching of carbon-martensitic steels may cause supersaturation of free carbon in the matrix, which decreases toughness. However, it was felt that if the toughness of HP 9-4-30 (with 0.33 percent C) could be improved by multistep heat treatment, it might represent the upper carbon limit for improved toughness of carbon-martensitic steels.

A phase transformation diagram for HP 9-4-30 was not available; therefore the iron-nickel equilibrium and transformation diagrams (figs. 2 and 3) were used to select the required temperatures. The chemical analysis of the HP 9-4-30 material (table II) showed a Ni content of 7.43 percent. This indicated that the grain refinement steps 2-5 should be raised approximately 50°F above the values used for HP 9-4-20. The annealing temperature of 1650°F and the tempering temperature of 1050°F were retained. This heating and cooling schedule is shown in figure 16 and is designated GR 30-I.

The as-received material consisted of a needle-like martensitic lath structure with an average grain size of 40-50 μm (ASTM size 6). Typical structure of the as-received material is shown in figure 17.

In the first specimen subjected to the GR 30-I heat treatment, the needle-like structure was transformed into a more equiaxed martensitic structure similar in appearance to the HP 9-4-20 GR 20-I material, with a grain size of 10-15 μm. The hardness of this specimen was 31-32 R_c compared with 38 R_c for the control specimen. The microstructure of the GR 30-I specimen is shown in figure 18.

The second specimen was subjected to the same grain-refining schedule except the tempering cycle (step 6) was 900°F for 1 hour. The microstructure produced by this treatment (GR 30-II) was almost identical in appearance to that of the GR 30-I material. The grain size of the GR 30-II specimen was 10-15 μm (the same as the grain size of the GR 30-I specimen) with a hardness of 29-30 R_c. The lower tempering temperature did not improve the grain refinement or the tensile properties over those produced by the GR 30-I heat treatment. The microstructure in figure 19 is typical of the GR 30-II specimen.

Charpy impact specimens were fabricated from the GR 30-I and the control specimen for testing at both room and cryogenic temperatures. The control specimens displayed the poor machinability typical of HP 9-4-30, while the GR 30-I specimens appeared to machine easier but caused high tool wear. End mill cutters dulled at a faster rate on the GR 30-I material than on the control material, although smoother surfaces could be machined while the cutters lasted.

The Charpy impact energy at -320°F was 20 ft-lb with lateral expansion of 9 mils for the GR 30-I material and 8 ft-lb with lateral expansion of 3 mils for the control material. Charpy test data for the GR 30-I and the control specimens are summarized in table V.

Because of the machining difficulties encountered and the failure to achieve the minimum requirement of C_{vn} = 25 ft-lb, no further experiments were performed on HP 9-4-30.

AF-1410 Pseudo-Maraging Steel

This alloy is a further development of the HY-180 advanced submarine hull steel containing a nominal 14 percent Co and 10 percent Ni. AF-1410 achieves its high tensile strength (230-240 ksi) through a combination of martensite tempering and secondary age hardening. Because of this dual strengthening mechanism, it is often referred to as a pseudo-maraging steel.

Although this steel possesses excellent toughness at room temperature, tests at Langley had revealed a very low (6-8 ft-lb) Charpy impact energy at -320°F. An attempt was made to improve the impact energy through grain refining. It was felt that this alloy with its complicated hardening system would provide useful information for the subsequent experiments with the 18Ni 200 grade fully maraging steel.

The only AF-1410 material available for grain refining consisted of standard 10-mm Charpy specimens cut from 5/8-in. plate stock and heat treated as shown in table I. A seven-step heating and cooling schedule was devised as follows: step 1, annealing to remove existing heat treatment; steps 2-5, cycles to refine grain; step 6, austenitizing to rehomogenize precipitates; and step 7, age hardening to restore strength. A phase diagram was not available for this alloy. The equilibrium and transformation diagrams (figs. 2 and 3) indicated grain-refining temperatures of approximately 1175°F and 1325°F. Because of the high Co content (13.95 percent), these temperatures were increased to 1200°F and 1350°F. The standard annealing and austenitizing temperatures of 1650°F and 1500°F were retained. This heating and cooling schedule is shown in figure 20 and is designated GR 1410.

Four Charpy specimens were subjected to the GR 1410 heat treatment. The hardness of these specimens was 46-48 R_c compared with 50-52 R_c for the standard heat-treated (control) specimens. After the GR 1410 heat treatment, the specimens were inspected for dimensional accuracy and then tested along with four control specimens at -320°F. No attempt was made to polish the notches or remove the light oxide film from the grain-refined specimens. The control specimens, machined from the stock plate material, were ground to a high-quality finish. Charpy impact energy of the GR 1410 specimens averaged 12 ft-lb compared with 7 ft-lb for the control material (see table VI).

After testing, metallographic specimens were cut from the ends of both the GR 1410 and the control Charpy specimens for microstructural comparison. The microstructure of both samples consisted of a fine-grained martensite lath structure with poorly defined grain boundaries. Actual grain size could not be measured because of the lack of grain boundary network detail. There was very little difference in appearance between the GR 1410 and the control specimens. The GR 1410 material appeared to have a more ordered and uniform subgrain structure, but actual reduction of grain size could not be determined (see fig. 21). The increased Charpy impact energy at -320°F could not be attributed to major alteration of the microstructure as observed in the 9Ni and HP alloys.

Hardness measurements made after the austenitizing cycle (step 6, fig. 20) and after the age hardening cycle (step 7) were 40 R_c and 46-48 R_c, respectively. The secondary age hardening mechanism was not impaired by the grain-refining heat treatment. This observation formed the basis for the next phase of experiments with a fully maraging steel (18Ni 200 grade) which would be annealed and age hardened after the grain refinement cycles.

18Ni 200 Grade Fully Maraging Steel

The 18Ni maraging steels are especially attractive for use in highly loaded wind-tunnel models because of their exceptional tensile strength (200-350 ksi) and ease of fabrication. Most maraging steels, however, lack sufficient toughness at cryogenic temperatures to be seriously considered for use in cryogenic wind tunnels. Only the 200-ksi grade offered any promise of meeting the required Charpy impact energy of 25 ft-lb at -320°F. Charpy energies of 25-30 ft-lb, which have been observed at -320°F in 18Ni 200 grade for specimens of small cross section (approx. 1/2 in. thick), are widely referenced in the literature of various manufacturers. Initial tests conducted at Langley consistently produced values of 14-17 ft-lb at -320°F for specimens taken from 1- to 5-in-thick plates and heavy bar stock. Therefore, experiments were performed to determine whether the cryogenic toughness could be improved significantly through grain refining.

Saul, Roberson, and Adair (ref. 12) have reported considerable reduction in the grain size of 250 and 300 grade maraging steel subjected to multiple heating cycles well above the austenitizing temperature in the range from 1700°F to 1880°F. Large grain size, 200 μm (ASTM size 2), was successfully reduced to 40 μm (ASTM size 7) with this high-temperature technique. The approach employed in this investigation for 18Ni 200 grade consisted of multiple heating and cooling cycles below the austenitizing temperature in the dual-phase region, similar to the procedure used for the other materials.

The as-received material consisted of annealed nickel martensite with bulky irregularly shaped grains of 60 to 100 μm. The hardness of the as-received material was 28 R_c. After the material was age hardened at 900°F for 3 hours, its hardness increased to 43 R_c with the structure becoming more ordered and lath-like in appearance with little if any change in grain size. The micrographs in figure 22 are typical of the as-received and age-hardened material.

It was assumed that if a suitable heating range could be found for grain refining, it would probably be narrow (150-200°F), requiring close control of specimen temperatures. The iron-nickel transformation diagram (fig. 3) was used to estimate an approximate temperature range from 1150°F to 1300°F. The exact temperature range was then determined by trial and error using 4 × 4 in. samples sawed from one of the 12 × 12 in. specimens.

The first attempt at grain refining consisted of multiple heating and cooling cycles at 1300°F followed by annealing at 1500°F to redesolve precipitates and age hardening at 900°F. This schedule was designated GR-1V and is shown in figure 23. Hardness values measured after the anneal cycle (step 5) averaged 26 R_c compared with 28 R_c for the annealed control specimen. After age hardening of the GR-1V material, its hardness measured 40 R_c compared with 43 R_c for the aged control material. Micrographs of the GR-1V specimen revealed no change in grain size or appearance compared with the control specimen.

The second attempt used multiple heating and cooling cycles at 1150°F with subsequent annealing and age hardening. This schedule (GR-2V) is shown in figure 24. The hardness values produced by this heat treatment were 28 R_c annealed and 44 R_c aged, essentially unchanged from the control material. The grain structure of this material was similar in shape and appearance to that of the control material, but with the grain size reduced approximately 25 percent (50 to 75 μm).

The third attempt used alternating cycles of 1250°F and 1100°F, followed by annealing and age hardening. This schedule was designated GR-3V and is shown in figure 25. The hardness values for the GR-3V material were 28 R_c annealed and 44 R_c aged. Micrographs of the structure revealed a significant reduction in grain size. The specimen consisted of a uniform lath-like structure with a grain size of 5-10 μm. The micrographs in figure 26 are typical of the GR-3V material. This degree of grain refinement is similar to the results obtained for HP 9-4-20.

The fourth attempt used the same heating and cooling schedule as for specimen GR-3V with the anneal cycle (step 5) changed to 1600°F. This schedule (GR-4V) is shown in figure 27. The hardness values produced by this treatment were 26 R_c annealed and 44 R_c aged. The structure consisted of very fine grains (5-10 μm); however, the grains were more blocky and irregular in shape than the uniform lath structure observed in the GR-3V specimen. The micrographs in figure 28 are typical of the GR-4V material.

The two remaining 12 × 12 in. plates were subjected to the GR-3V and GR-4V heat treatments through the annealing cycle (step 5). Charpy impact specimens were then fabricated from the GR-3V, the GR-4V, and the control specimen for testing at room temperature and -320°F. The Charpy specimens were machined in the annealed condition and then age hardened at 900°F. After age hardening, the specimens were tested without any polishing or removal of the oxide film. During the machining operation, Langley fabrication personnel characterized the grain-refined specimens as easier to machine than the control specimens. Higher removal rates, smoother surface finishes, and longer tool life were observed for the GR-3V and GR-4V materials.

The Charpy impact energy at -320°F averaged 24.0 ft-lb for GR-3V, 20.2 ft-lb for GR-4V, and 15.8 ft-lb for the control. Lateral expansions were 4, 3, and 3 mils, respectively. A complete summary of the Charpy data is shown in table VII.

Additional experiments were conducted using the GR-3V heat treatment with various age hardening temperatures (step 6) in an attempt to reach the program goal of C_{vn} = 25 ft-lb. Two additional 4 × 4 in. specimens were subjected to the GR-3V treatment through the annealing cycle (step 5). One specimen was then aged for 3 hours at 1000°F and the other for 3 hours at 1100°F. These heat treatments were designated GR-3V-1000 and GR-3V-1100.

The grain structure of the GR-3V-1000 specimen consisted of very fine grains of 5-7 μm with a more uniform appearance than the structure of the GR-3V specimen. The micrographs in figure 29 are typical of the GR-3V-1000 specimen. The hardness of this sample was 43-44 R_c.

The microstructure of the GR-3V-1100 specimen was more disordered than the structure of the GR-3V specimen, with grain sizes ranging from 10 to 30 μm. The grains appeared more blocky and irregular in shape than the lath structure produced by the GR-3V heat treatment. The hardness of this sample was 39 R_c. The 1100°F age cycle was apparently detrimental to both the grain refinement and the age hardening response. The microstructure of the GR-3V-1100 material is shown in figure 30.

Charpy specimens were then cut from the annealed GR-3V material and age hardened at 1000°F. The impact strength at -320°F was 20 ft-lb with lateral expansion of 8 mils. These data are shown in table VII.

The GR-3V material, age hardened at 900°F, produced the best combination of strength and impact strength although it just fell short of meeting the goal of

C_{vn} = 25 ft-lb at -320°F (the average value was 24 ft-lb). An additional set of four GR-3V and four control Charpy specimens were prepared and tested at -275°F. This temperature was chosen because it is considered a typical low operating temperature for most models to be tested in the NTF. The specimens were submerged for 1 hour in liquid freon cooled to -275°F by a LN_2 heat exchanger. The specimens were removed and tested within 5 seconds. The average impact energies were 26.5 ft-lb for the GR-3V specimen and 16.3 ft-lb for the control specimen. These data are also shown in table VII.

Fractographs were made of the -320°F Charpy fracture surfaces of the control, GR-3V, GR-4V, and GR-3V-1000 materials. The fracture mode of all four specimens appeared to be microvoid coalescence with a larger number of small fracture sites visible on the grain-refined surfaces. There was more extensive plastic straining and dimpling between microvoids on all grain-refined specimens than on the control specimen. These fractographs are shown in figure 31.

Tensile tests were performed on the control and GR-3V materials at both room temperature and -320°F. Yield and ultimate strengths were 262 and 273 ksi, respectively, for the GR-3V material at -320°F. The elongation as measured by crosshead travel for the GR-3V material was 0.20 in. compared with 0.08 in. for the control specimen at -320°F. Also, the GR-3V material showed improvement in all tensile properties over those of the control material. Tensile test data for the GR-3V and control specimens are summarized in table VIII.

GENERAL DISCUSSION OF RESULTS

Grain-Refining Heat Treatments

Multistep heat treatments to refine the grain size, developed for the ferritic steels, were extended to the quenched and tempered martensitic steels and the fully maraging steels. This technique resulted in grain size reductions of up to 1/10 of original size or smaller. The largest reductions were observed in the HP 9-4-20 and 18Ni 200 grade materials. The grain reduction process was demonstrated in alloys containing up to 0.33 percent C.

The grain-refining process requires multiple cycles between the austenitic (γ) region and the dual-phase ferrite plus austenite ($\alpha + \gamma$) region followed by rapid cooling. The critical parameter appears to be time at temperature for a given cycle and not heating or cooling rates. The transformations occur rapidly, on the order of tens of minutes, once the required temperature is reached, but the grain refinement can be erased if the heating cycle is held for several hours. This observation indicates a shear process as opposed to a diffusion process. The results indicate that the technique used in these investigations may be applicable to other alloy groups that possess a definitive $\alpha + \gamma$ region.

Cryogenic Toughness and Tensile Strength

The apparent toughness at -320°F as measured by Charpy impact energy was increased for every alloy investigated, although the increased toughness of AF-1410 could not be attributed to grain refinement. The indicated toughness increases for HP 9-4-20 of 180 percent and for 18Ni 200 grade of 52 percent at -320°F are exceptional, given the high strength of both alloys. The effect of grain refinement on the mechanical properties of all alloys used in this investigation are summarized in figure 32 (taken from ref. 3). Tensile properties were reduced from 0 to 25 percent for the various alloys tested. The data in figure 32 show that C_{vn} for the grain-refined materials increased more at -320°F than at room temperature for all alloys.

Grain refinement permitted HP 9-4-20 to exceed the program goal of C_{vn} = 25 ft-lb at -320°F, while 18Ni grade 200 fell just short of the goal. However the 18Ni grade 200 with C_{vn} = 24 ft-lb at -320°F together with its 146-percent relative increase in elongation and ultimate strength of 273 ksi is marginally acceptable. A program to fully characterize both HP 9-4-20 and 18Ni 200 grade grain-refined materials should be undertaken. This program should be designed to measure actual fracture toughness (K_{1c}) as well as both tensile and impact properties. Alloy phase stability and fabrication suitability should also be investigated. Studies should be made to optimize the heat treatment schedule for the best combination of strength and toughness available from the commercial grade materials.

CONCLUSIONS AND RECOMMENDATIONS

A program to improve the cryogenic toughness of high strength commercial steels through grain-refining heat treatments was conducted at Langley Research Center and is described in this paper. The following conclusions were reached:

1. Multistep heat treatments can be used to improve the cryogenic toughness of martensitic and maraging steels by reduction of grain size.

2. HP 9-4-20 steel exhibited the largest increase in toughness (approx. 180 percent) while retaining good medium range strength. It should receive strong consideration as a cryogenic model material for the strength requirement range from 100 to 150 ksi.

3. The grain-refined 18Ni 200 grade steel exhibited a combination of strength and toughness not obtained at Langley before in a single alloy. This alloy is currently being used for cryogenic models, and grain refinement should be considered for toughness enhancement. Charpy impact energy was increased by 52 percent with a slight increase in tensile properties at -320°F.

4. The machinability of all grain-refined alloys was judged to be improved over that of the stock alloys.

5. The results of the grain refinement investigation suggest that the techniques may be used for other alloy groups that possess a definite dual-phase ferrite plus austenite ($\alpha + \gamma$) region. Additional work to further characterize the transformation mechanism should be undertaken.

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National Aeronautics and Space Administration
Hampton, VA 23665
July 7, 1984

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TABLE I.- SPECIMEN SIZES, HEAT TREATMENT, AND ROCKWELL HARDNESS PRIOR TO GRAIN REFINEMENT

Alloy	Specimen size	Heat treatment prior to grain refinement (a)	Average hardness, R_c
9Ni (ASTM A353-79)	12 × 12 × 3/4 in. plate stock	1650°F/1 hr/A.C. 1450°F/1 hr/A.C. 1100°F/1 hr/W.Q.	21
HP 9-4-20	12 × 12 × 1 in. plate stock	1650°F/1 hr/W.Q. 1500°F/1 hr/W.Q. 1025°F/6 hr/W.Q.	39
HP 9-4-30	12 × 12 × 1 in. plate stock	1700°F/1 hr/A.C. 1550°F/1 hr/O.Q. 1100°F/4 hr/A.C.	38
AF-1410	10 × 10 × 55 mm Charpy specimens cut from 5/8-in. plate stock	1650°F/30 min/O.Q. 1500°F/30 min/O.Q. 950°F/5 hr/A.C.	52
18Ni 200 grade	12 × 12 × 1.3 in. plate split sawed from 4 1/2 × 26 1/2 × 95 1/2 in. forged plate	Annealed 1500°F/4 1/2 hr/A.C.	28

[a]A.C. = Air cooled, W.Q. = Water quench, O.Q. = Oil quench.

TABLE II.- CHEMICAL ANALYSIS OF ALLOYS

Alloy	Percent by weight of -																			
	C	Mn	Si	S	P	Cr	Ni	Co	Mo	Al	Ti	Cu	V	N	O	W	B	Zr	Ca	Fe
9Ni	0.07	0.448	0.23	0.005	0.013		8.90													Bal.
HP 9-4-20	.190	.37	.05	.003	.008	0.80	9.15	4.53	1.00			0.11	0.08							Bal.
HP 9-4-30	.330	.27	.06	.003	.008	1.10	7.43	4.50	1.07			.35	.10							Bal.
AF-1410	.16	.08	.03	.002	.004	1.99	10.20	13.95	1.04	0.003	0.005			0.0005	0.0005					Bal.
18Ni 200 grade	.006	.02	.04	.004	.004	.07	18.38	8.56	3.36	.13	.25	.10				0.02	0.003	0.010	0.05	Bal.

TABLE III.- CHARPY IMPACT TEST RESULTS FOR 9Ni ALLOY

[All data are averages of a minimum of four tests]

Specimen	Impact energy, C_{vn}, ft-lb		Lateral expansion, mils		Hardness, R_c	Change[a] in tensile strength from control value, percent
	Room temp.	-320°F	Room temp.	-320°F		
Control	124	73	84	45	21	
GR I	139	102	82	58	23	+2

[a]Estimated from R_c hardness using ASTM A 370 (ref. 9).

TABLE IV.- TEST RESULTS FOR HP-9-4-20

[All data are averages of a minimum of four tests]

(a) Charpy impact test results

Specimen	Impact energy, C_{vn}, ft-lb		Lateral expansion, mils		Hardness, R_c
	Room temp.	-320° F	Room temp.	-320° F	
Control	57.4	13.6	36	6	39
GR 20-I	88.6	38.5	56	20	32-33

(b) Tensile test results

Specimen	Yield strength, ksi		Ultimate strength, ksi		Elongation,[a] in.		Reduction in percent	
	Room temp.	-320° F	Room temp.	-320° F	Room temp.	-320° F	Room temp.	-320° F
Control	186.7	217.0	200.8	230.9	0.140	0.121	69.3	56.2
GR 20-I	118.6	160.5	158.7	224.6	.314	.345	60.5	40.8

[a]Elongation measured from crosshead advance at fracture.

TABLE V.- CHARPY IMPACT TEST RESULTS FOR HP 9-4-30

[All data are averages of a minimum of four tests]

Specimen	Impact energy, C_{vn}, ft-lb		Lateral expansion, mils		Hardness, R_c	Change[a] in tensile strength from control value, percent
	Room temp.	-320°F	Room temp.	-320°F		
Control	38.3	8.0	16	3	38	
GR 30-1	59.5	20.3	39	9	31-32	-16

[a]Estimated from R_c hardness using ASTM A 370 (ref. 9).

TABLE VI.- CHARPY IMPACT TEST RESULTS FOR AF-1410 AT -320°F

[All data are averages of a minimum of four tests]

Specimen	Impact energy, C_{vn}, ft-lb	Lateral expansion, mils	Hardness, R_c	Change[a] in tensile strength from control value, percent
Control	7.0	1	50-52	
GR 1410	12.0	4	46-48	-13

[a]Estimated from R_c hardness using ASTM A 370 (ref. 9).

TABLE VII.- CHARPY IMPACT TEST RESULTS FOR 18Ni 200 GRADE

[All data are averages of a minimum of four tests]

Specimen	Impact energy, C_{vn}, ft-lb			Lateral expansion, mils			Hardness, R_C
	Room temp.	-320°F	-275°F	Room temp.	-320°F	-275°F	
Control	29.3	15.8	16.3	9	3	4	42-44
GR-3V	37.5	24.0	26.5	15	4	6	44-45
GR-4V	37.8	20.2		14	3		44
GR-3V-1000	34.5	20.0		15	8		43-44

TABLE VIII.- TENSILE TEST RESULTS FOR 18Ni 200 GRADE

[All data are averages of a minimum of four tests]

Specimen	Yield strength, ksi		Ultimate strength, ksi		Elongation,[a] in.		Reduction in area, percent	
	Room temp.	-320°F	Room temp.	-320°F	Room temp.	-320°F	Room temp.	-320°F
Control	200.5	248.6	204.6	255.8	0.10	0.08	38.0	30.4
GR-3V	205.0	262.0	210.6	273.1	.18	.20	56.7	53.5

[a]Elongation measured from crosshead advance at fracture.

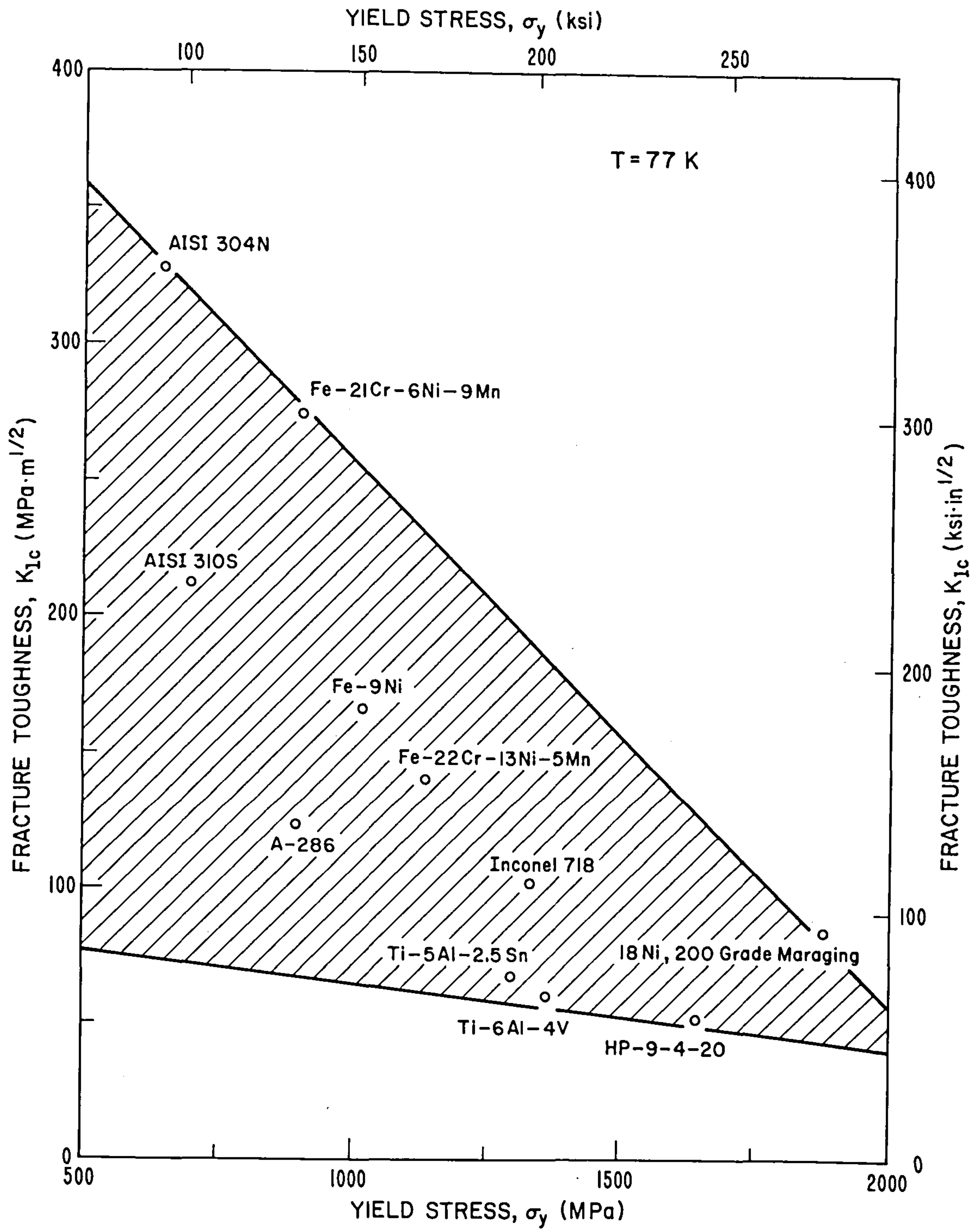


Figure 1.- Toughness-versus-strength trend for structural metals at -320°F. (From ref. 1.)

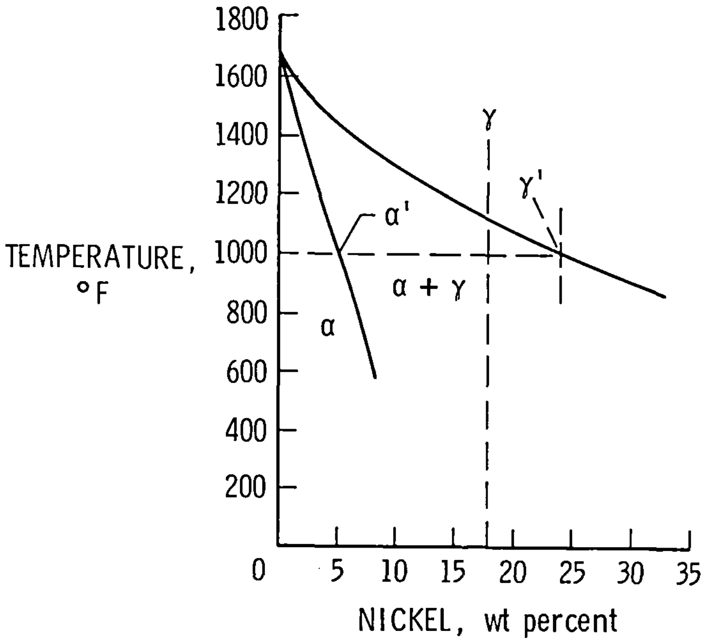


Figure 2.- Iron-nickel equilibrium diagram. (From ref. 8.)

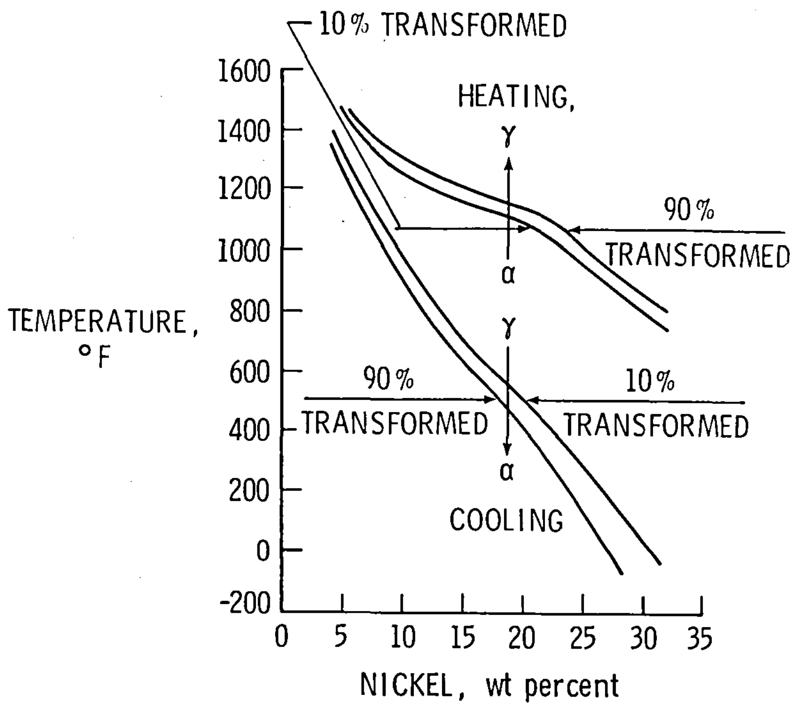


Figure 3.- Iron-nickel transformation diagram. (From ref. 8.)

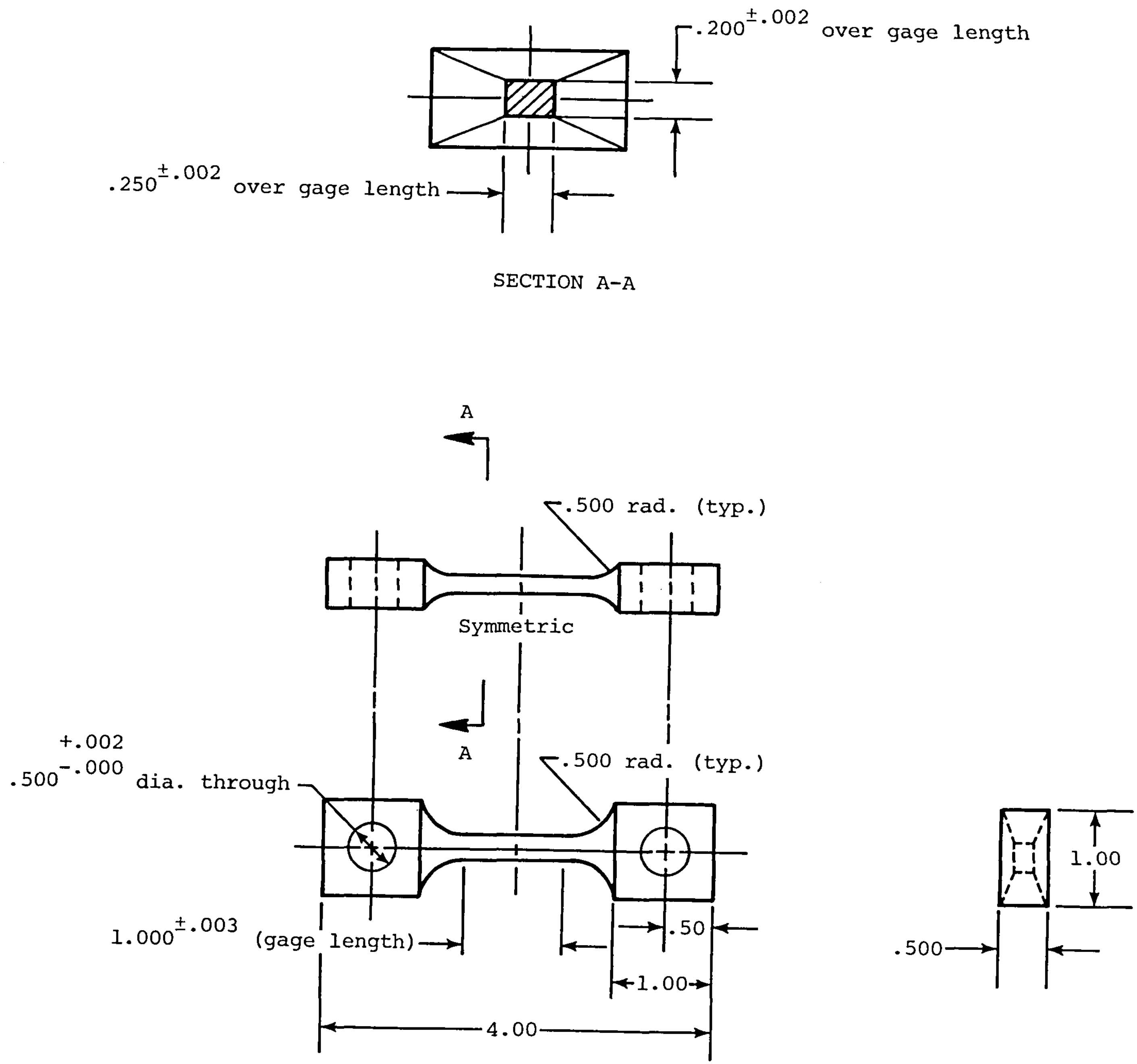
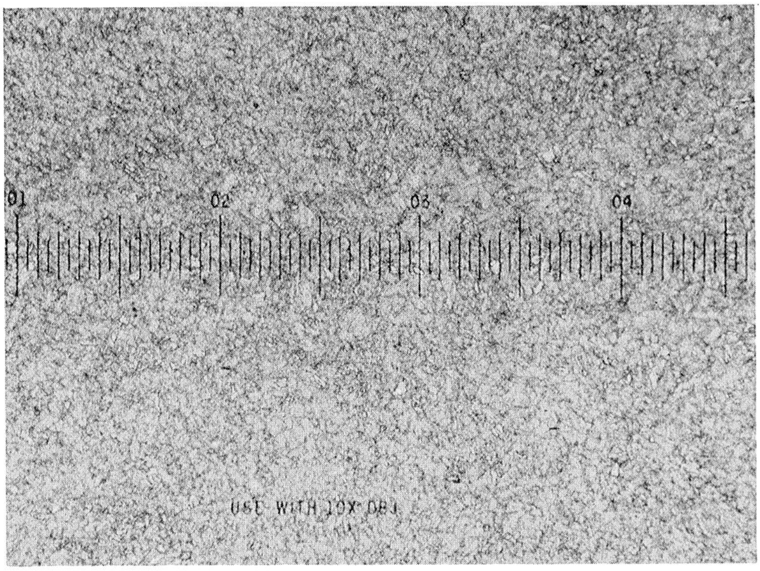
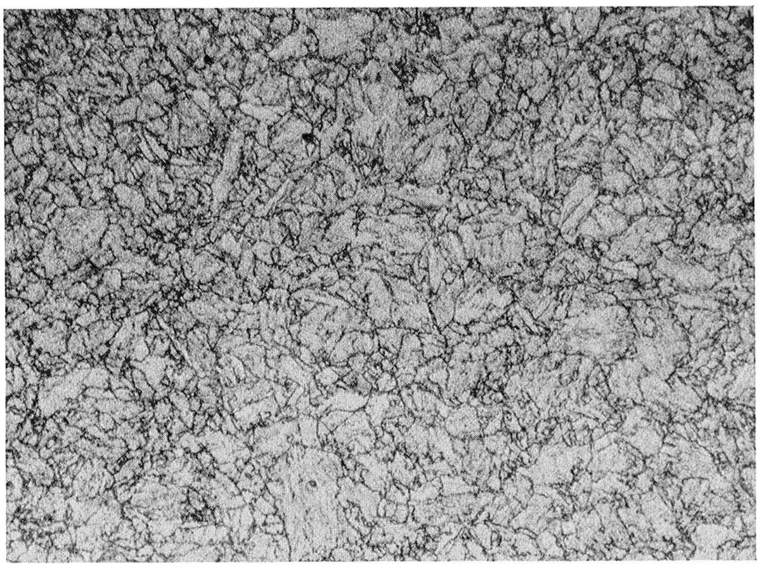


Figure 4.- Tensile specimen geometry. All dimensions are in inches.



100 µm

× 100



10 µm

× 400

L-84-79

Figure 5.- Microstructure of 9Ni control specimen.

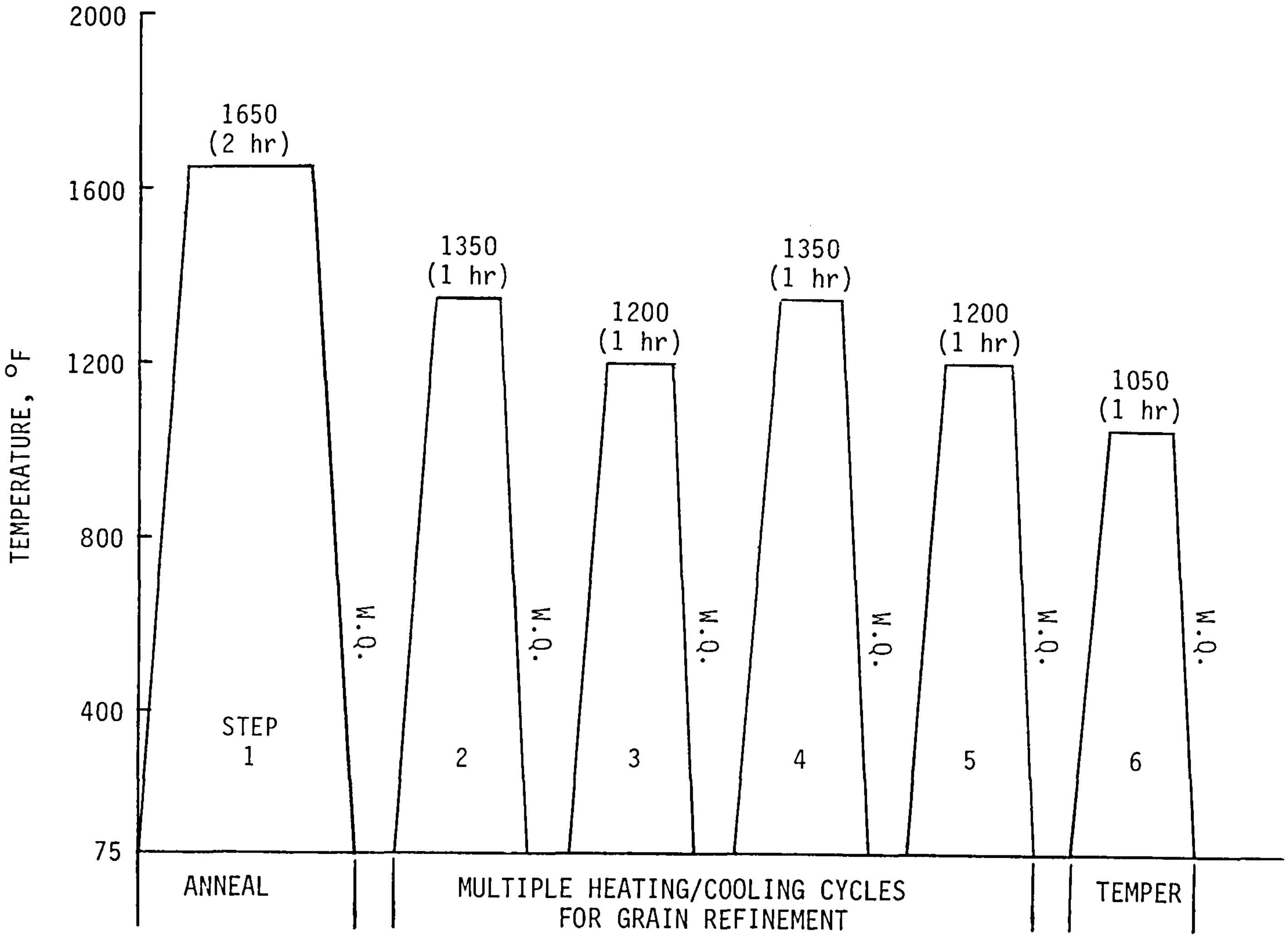
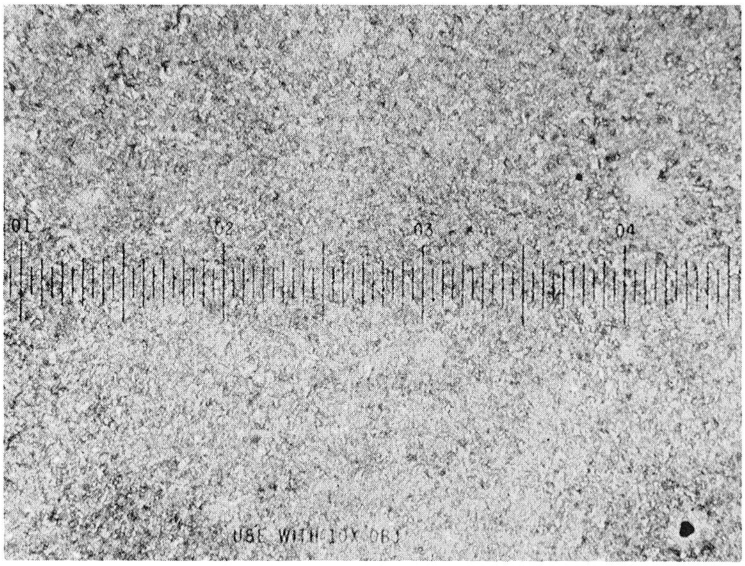
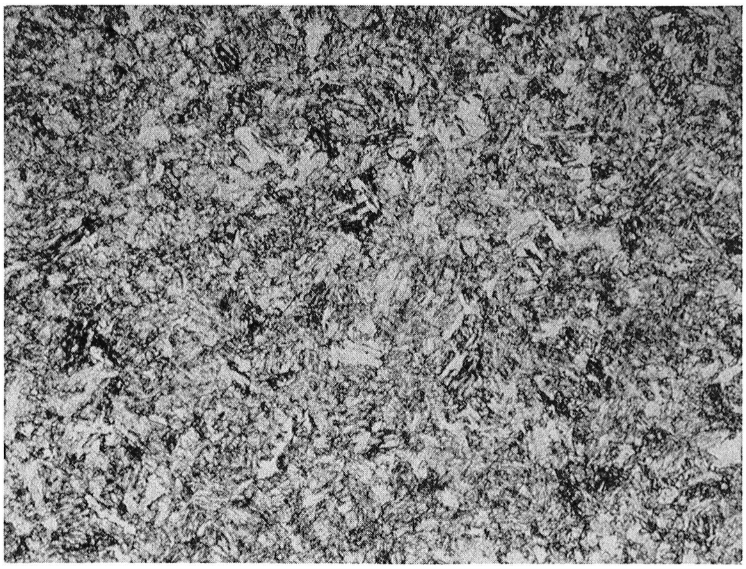


Figure 6.- Grain-refining heat treatment GR I for 9Ni.



100 µm

× 100

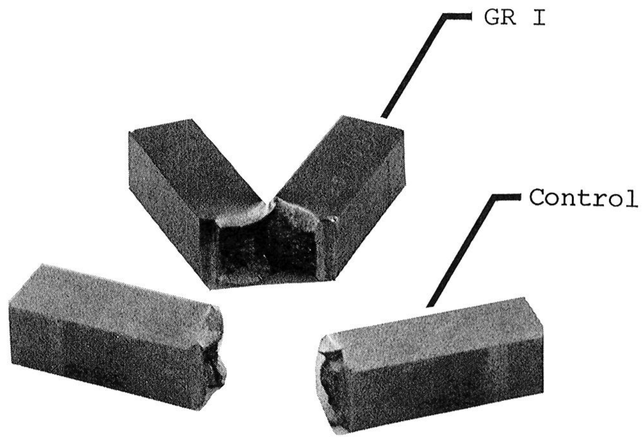


10 µm

× 400

L-84-80

Figure 7.- Microstructure of 9Ni GR I specimen.



GR I, C_{vn} = 114 ft-lb (incomplete fracture)

Control, C_{vn} = 67 ft-lb

L-83-4421

Figure 8.- 9Ni Charpy impact specimens tested at -320°F.

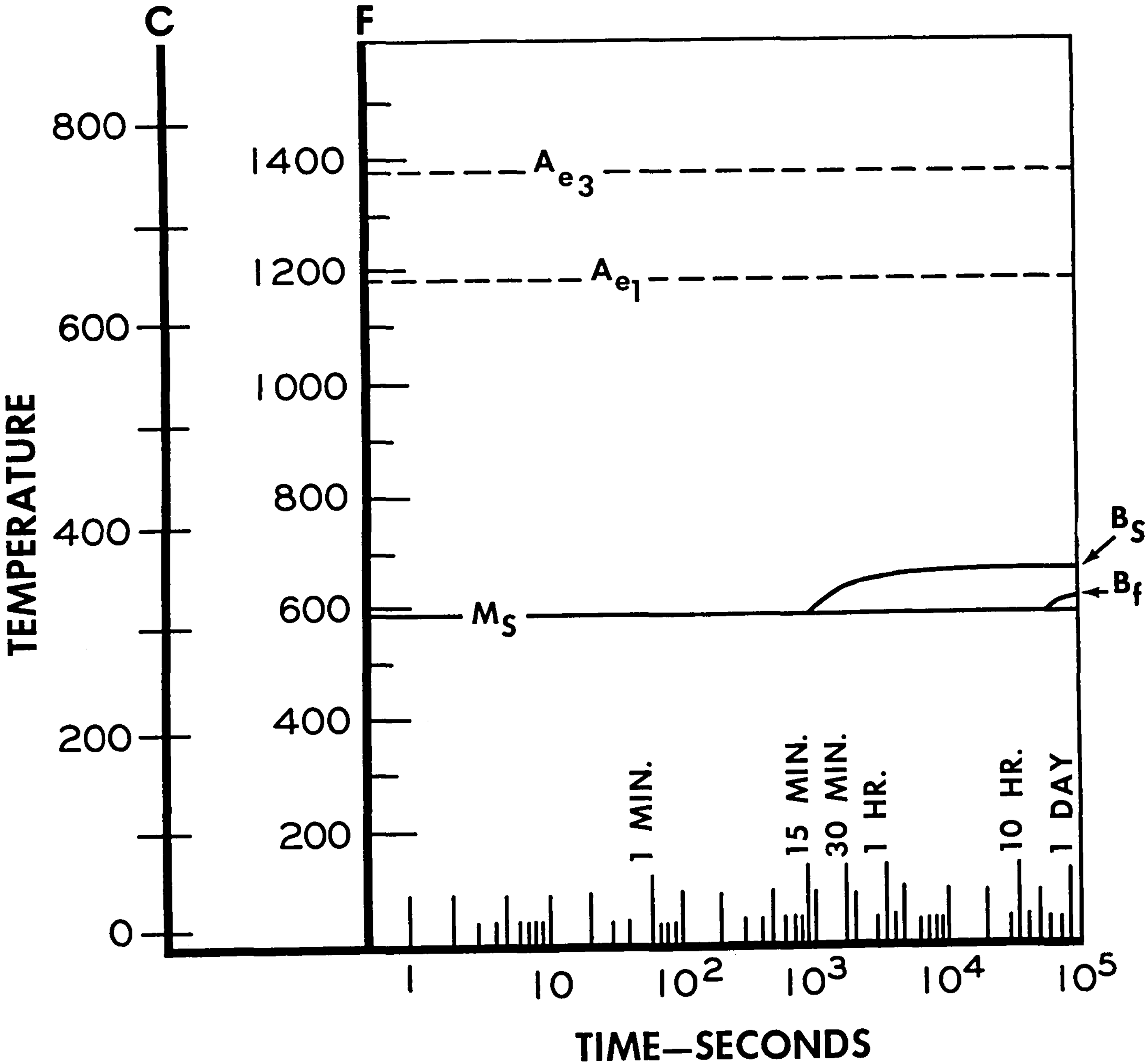


Figure 9.- Isothermal transformation diagram for HP 9-4-20 (Heat 3931677). (From ref. 10.)

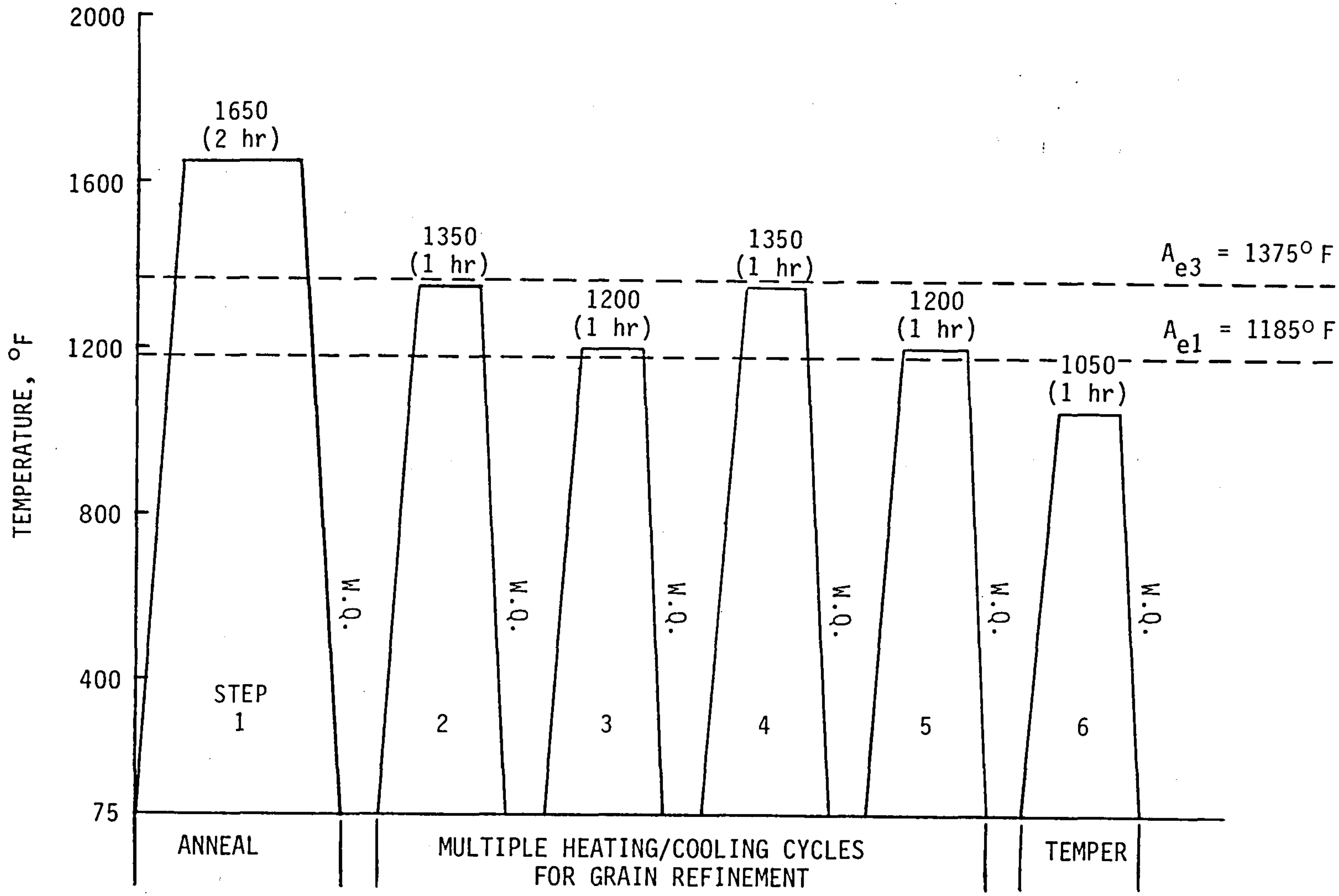
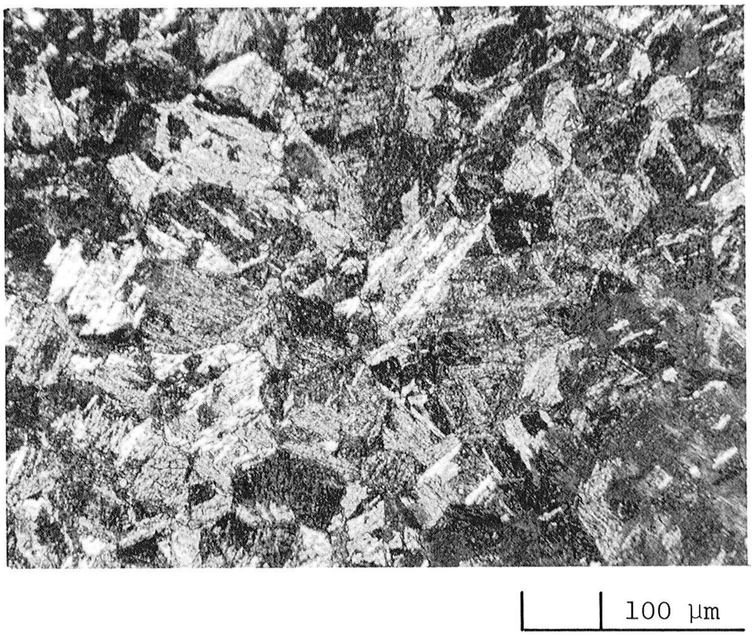
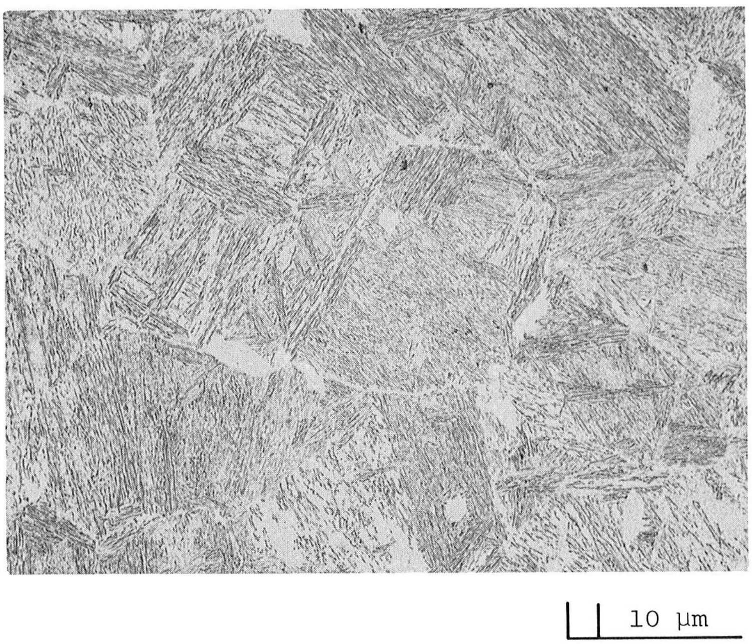


Figure 10.- GR I heat treatment with HP 9-4-20 phase change temperatures superimposed.



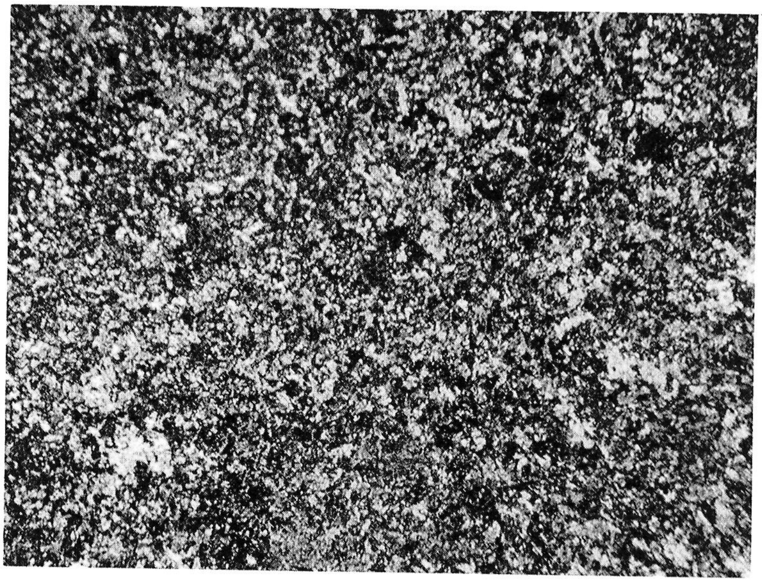
× 100



× 400

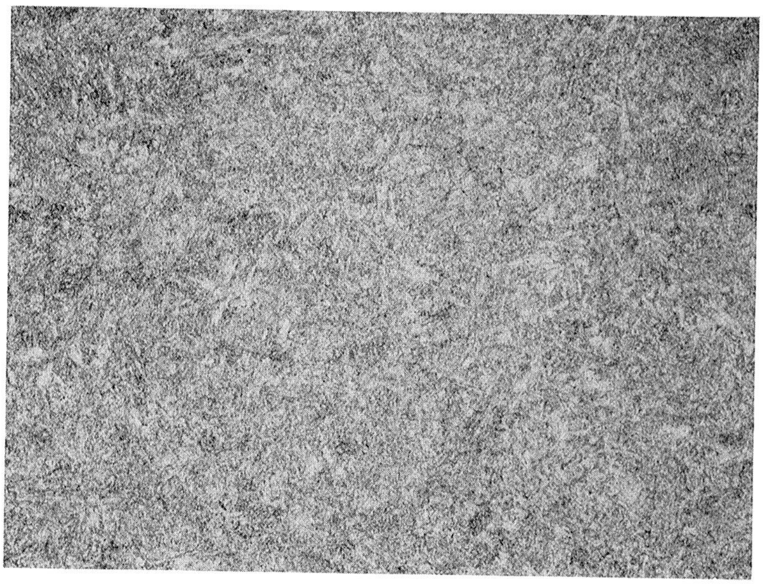
L-84-81

Figure 11.- Microstructure of HP 9-4-20 control specimen.



100 μm

× 100

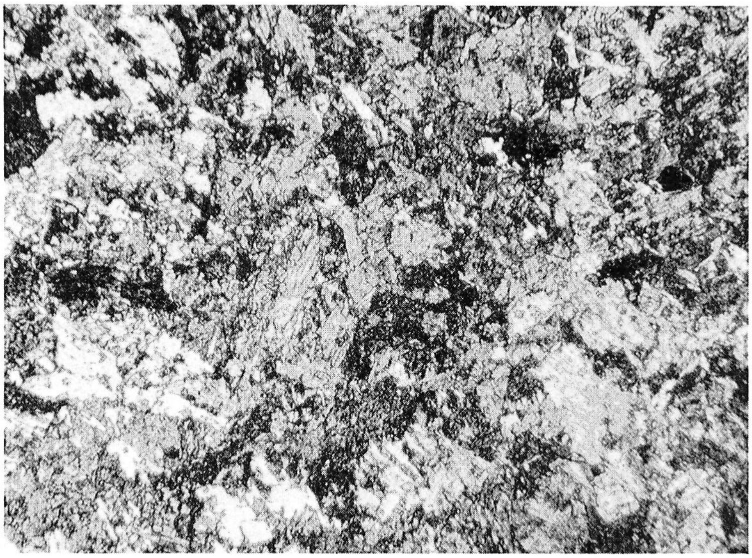


10 μm

× 400

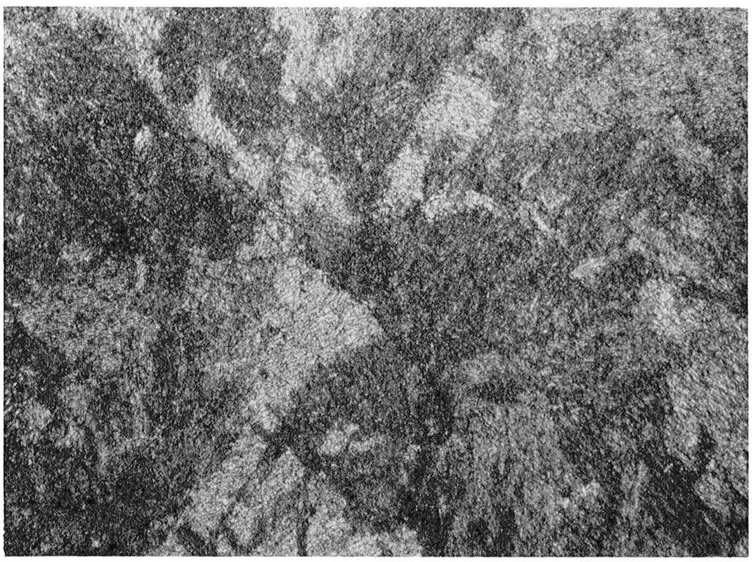
L-84-82

Figure 12.- Microstructure of HP 9-4-20 GR 20-I specimen.



100 μm

× 100

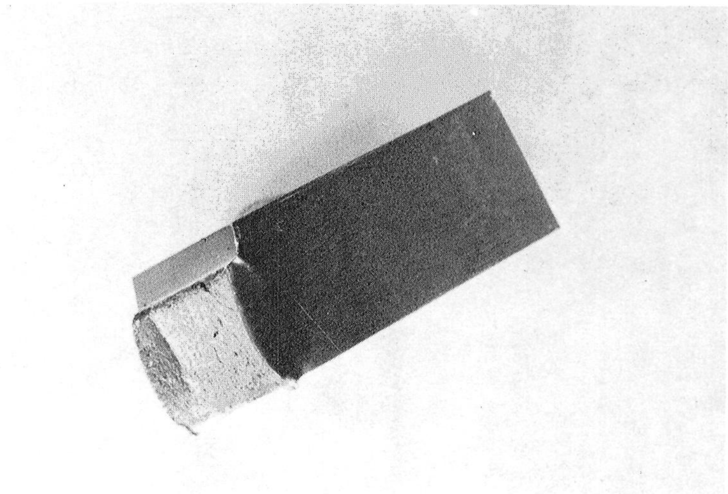


10 μm

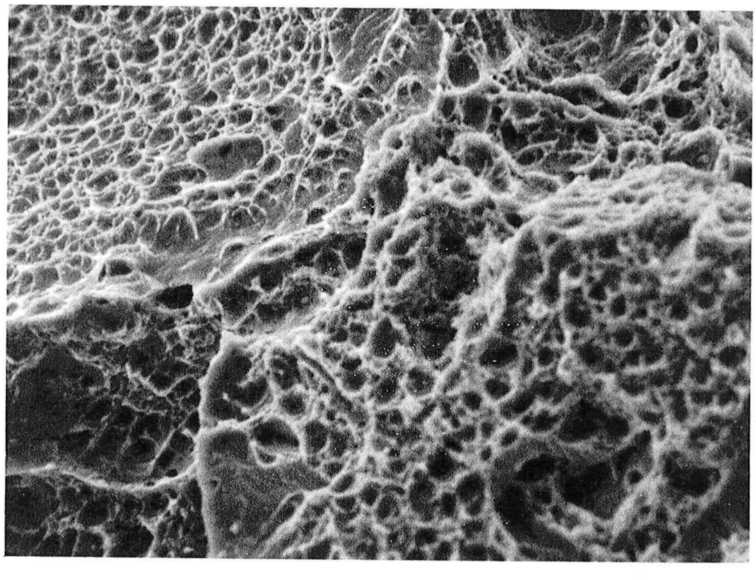
× 400

L-84-83

Figure 13.- Microstructure of HP 9-4-20 GR 20-II specimen.



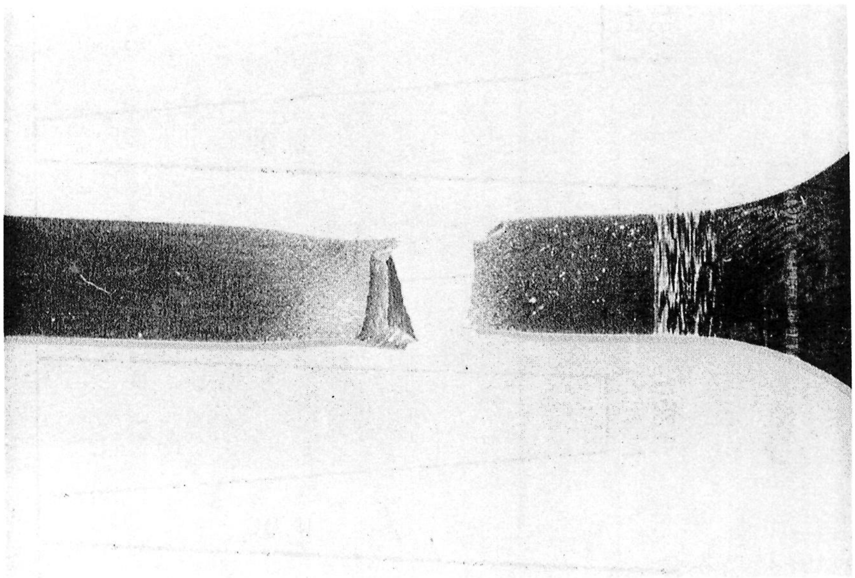
C_{vn} = 39 ft-lb, Lateral expansion = 20 mils



Fracture surface, × 2000

L-84-84

Figure 14.- HP 9-4-20 GR 20-I Charpy impact specimen tested at -320°F.



Ultimate strength = 223.4 ksi, Elongation = 0.339 in.

L-83-7144

Figure 15.- HP 9-4-20 GR 20-I tensile specimen tested at -320°F.

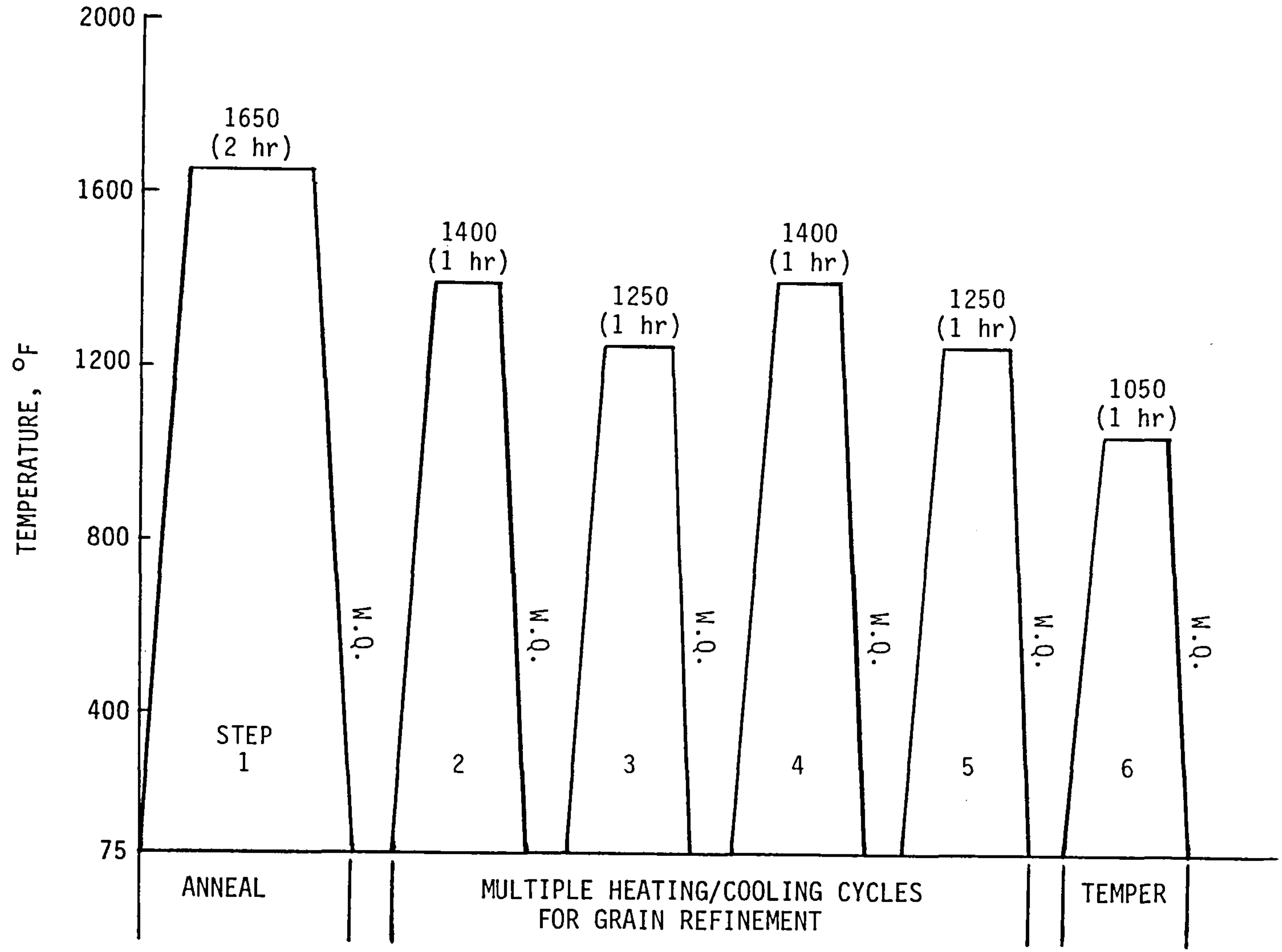
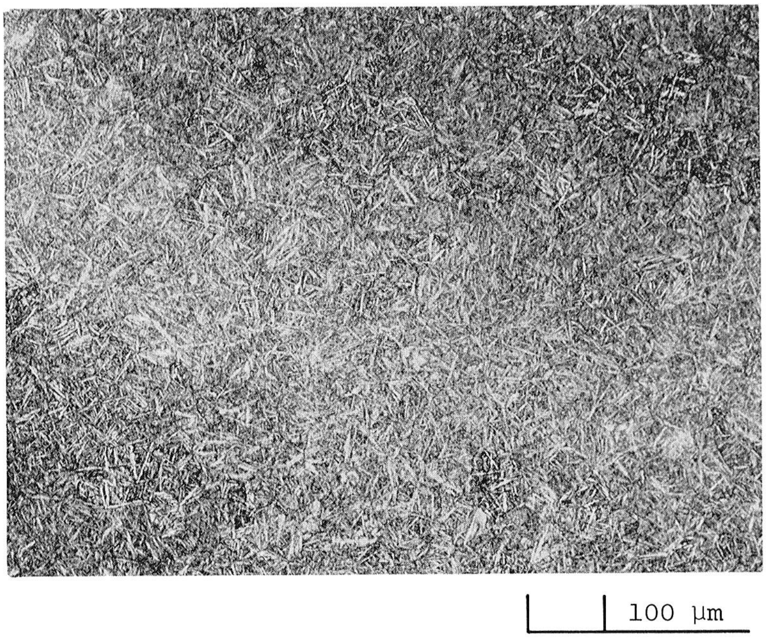
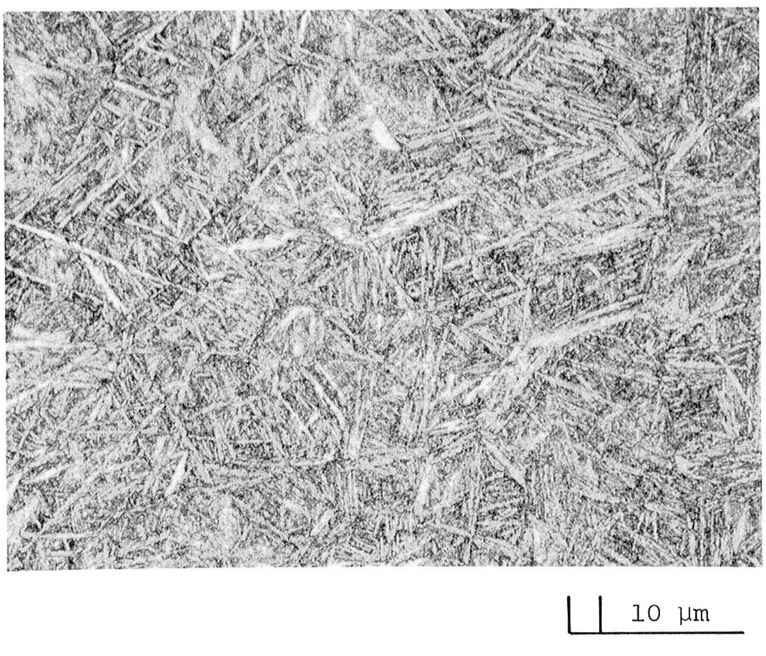


Figure 16.- Grain-refining heat treatment GR 30-I for HP 9-4-30.



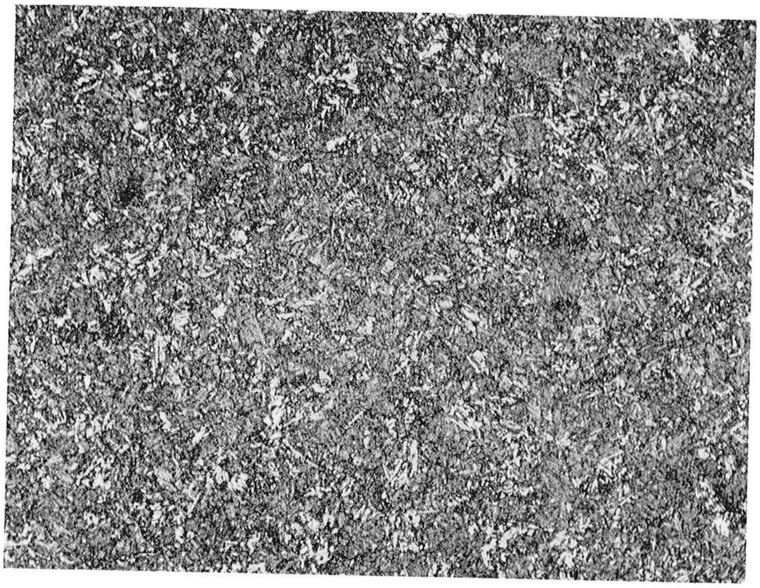
× 100



× 400

L-84-85

Figure 17.- Microstructure of HP 9-4-30 control specimen.



100 μm

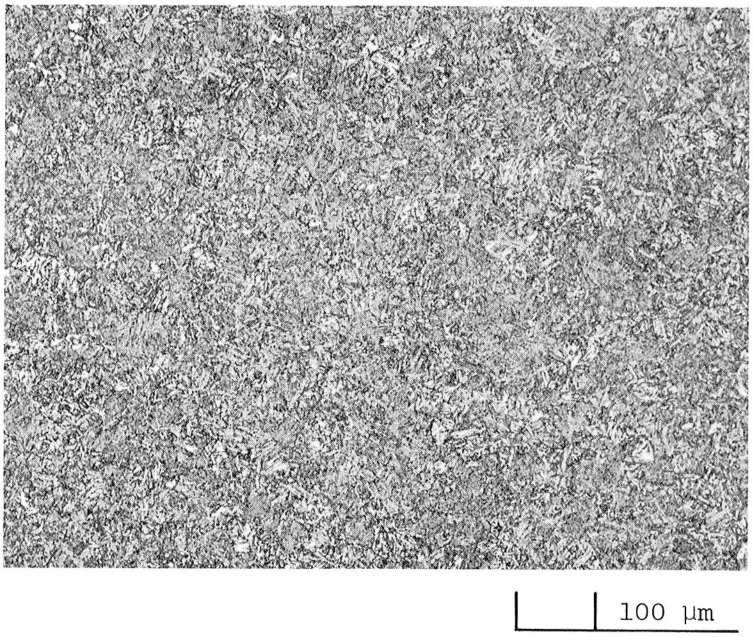
× 100

10 μm

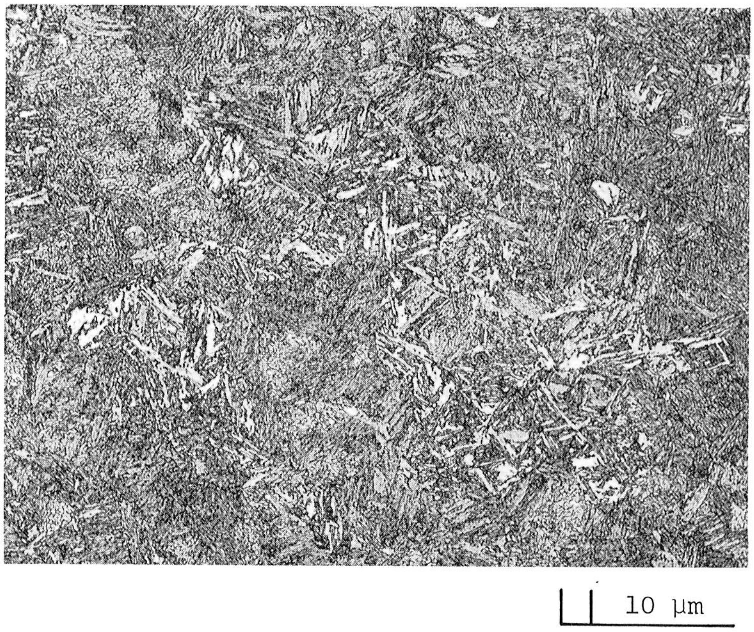
× 400

L-84-86

Figure 18.- Microstructure of HP 9-4-30 GR 30-I specimen.



× 100



× 400

L-84-87

Figure 19.- Microstructure of HP 9-4-30 GR 30-II specimen.

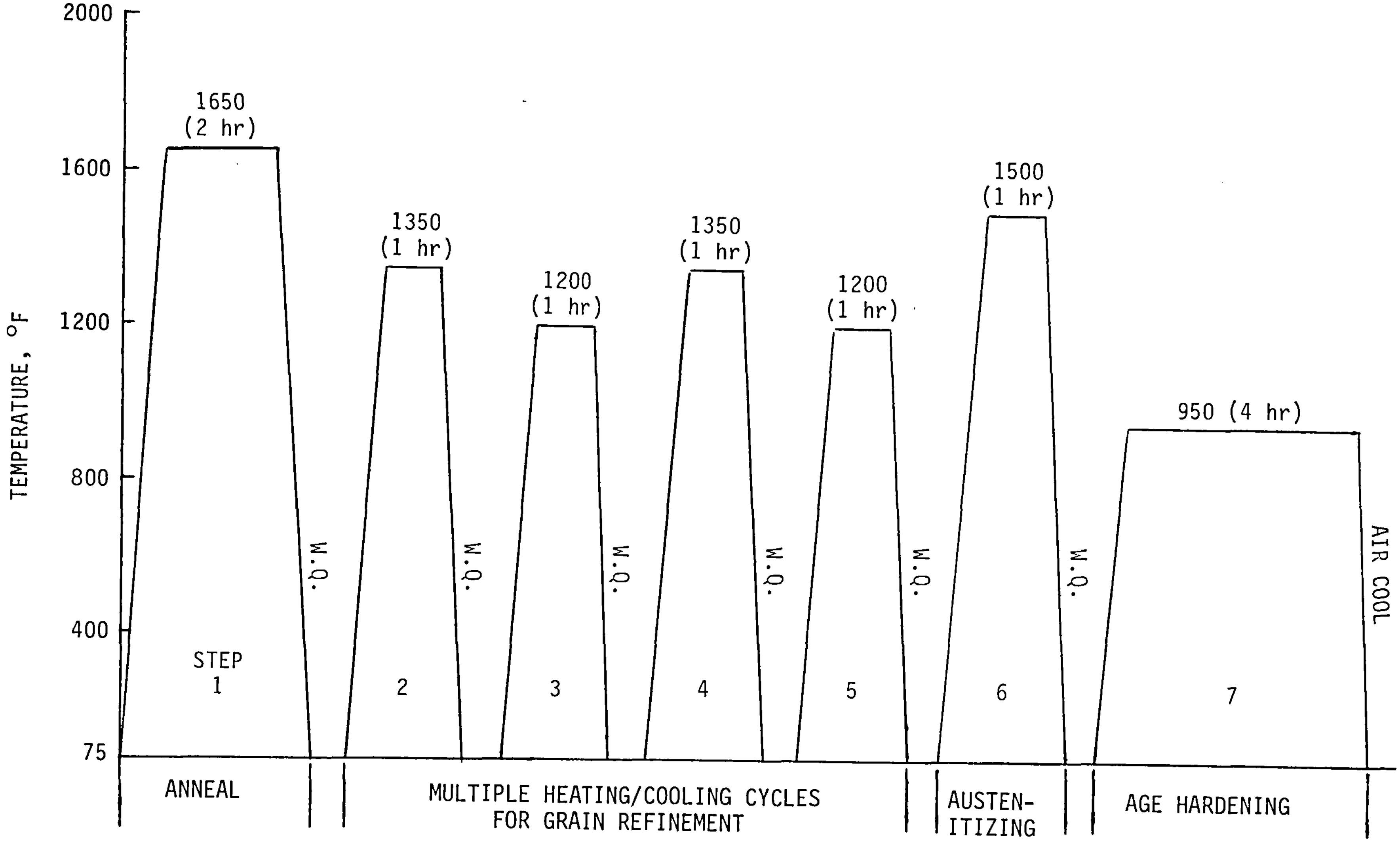
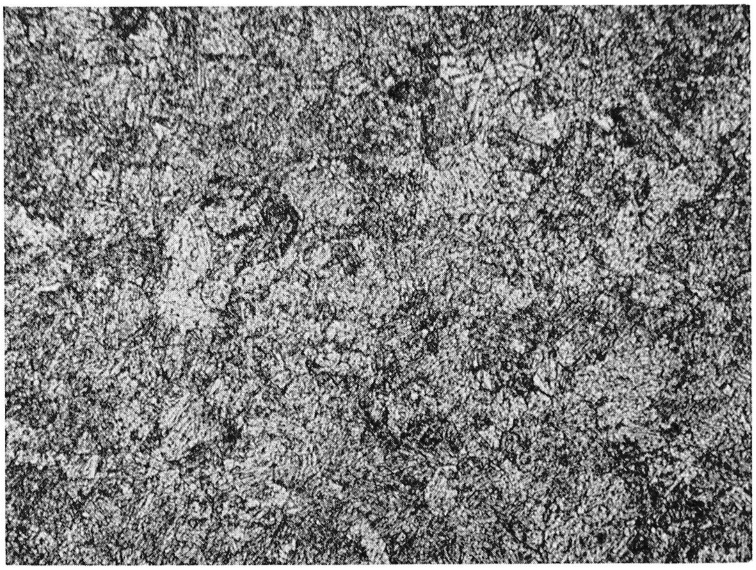
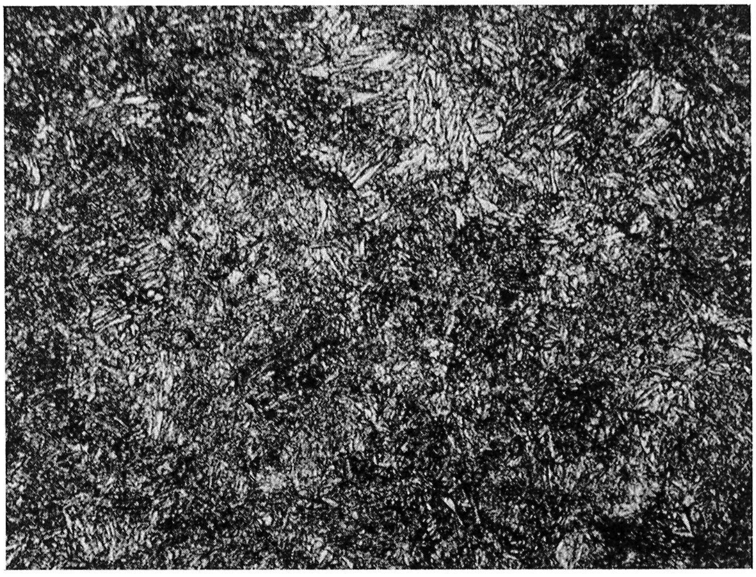


Figure 20.- Grain-refining heat treatment GR 1410 for AF-1410.



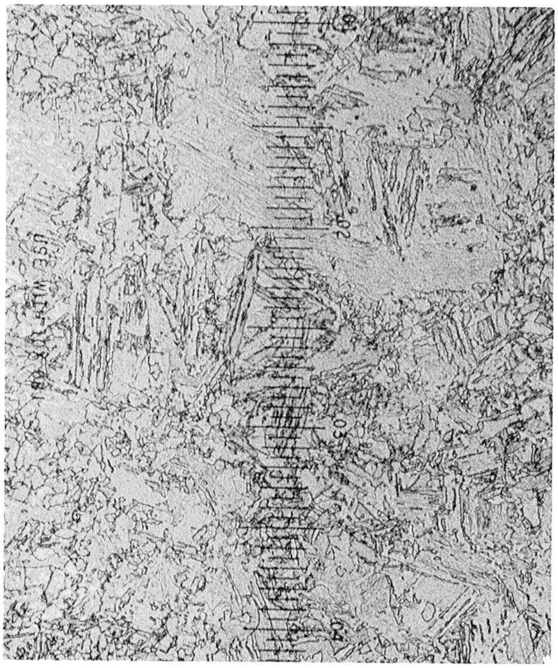
Control, × 400



GR 1410, × 400

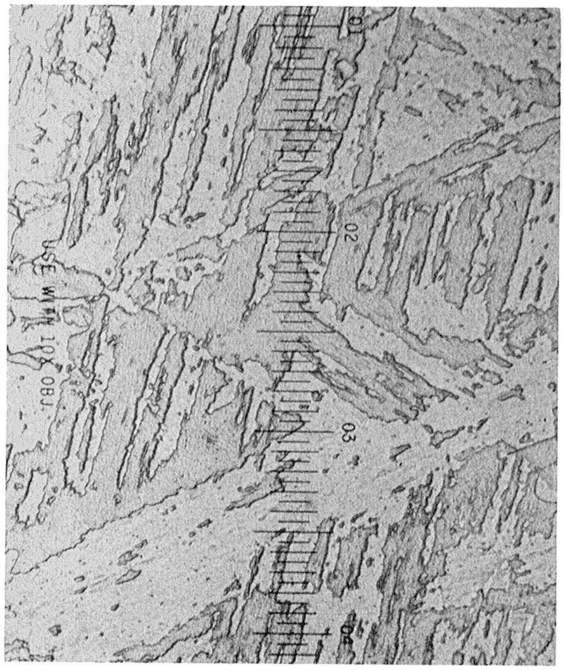
L-84-88

Figure 21.- Microstructure of AF-1410 specimens.



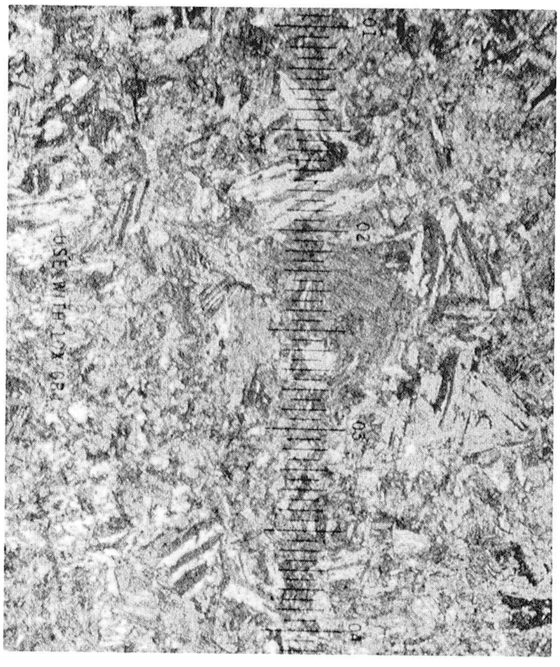
100 μm

Annealed, × 100



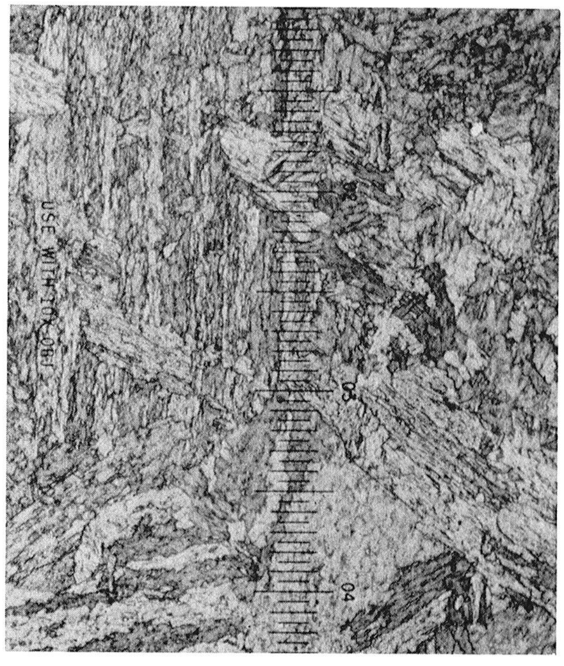
10 μm

Annealed, × 400



100 μm

Aged at 900°F, × 100



10 μm

Aged at 900°F, × 400

L-84-89

Figure 22.- Microstructure of 18Ni 200 grade control specimens.

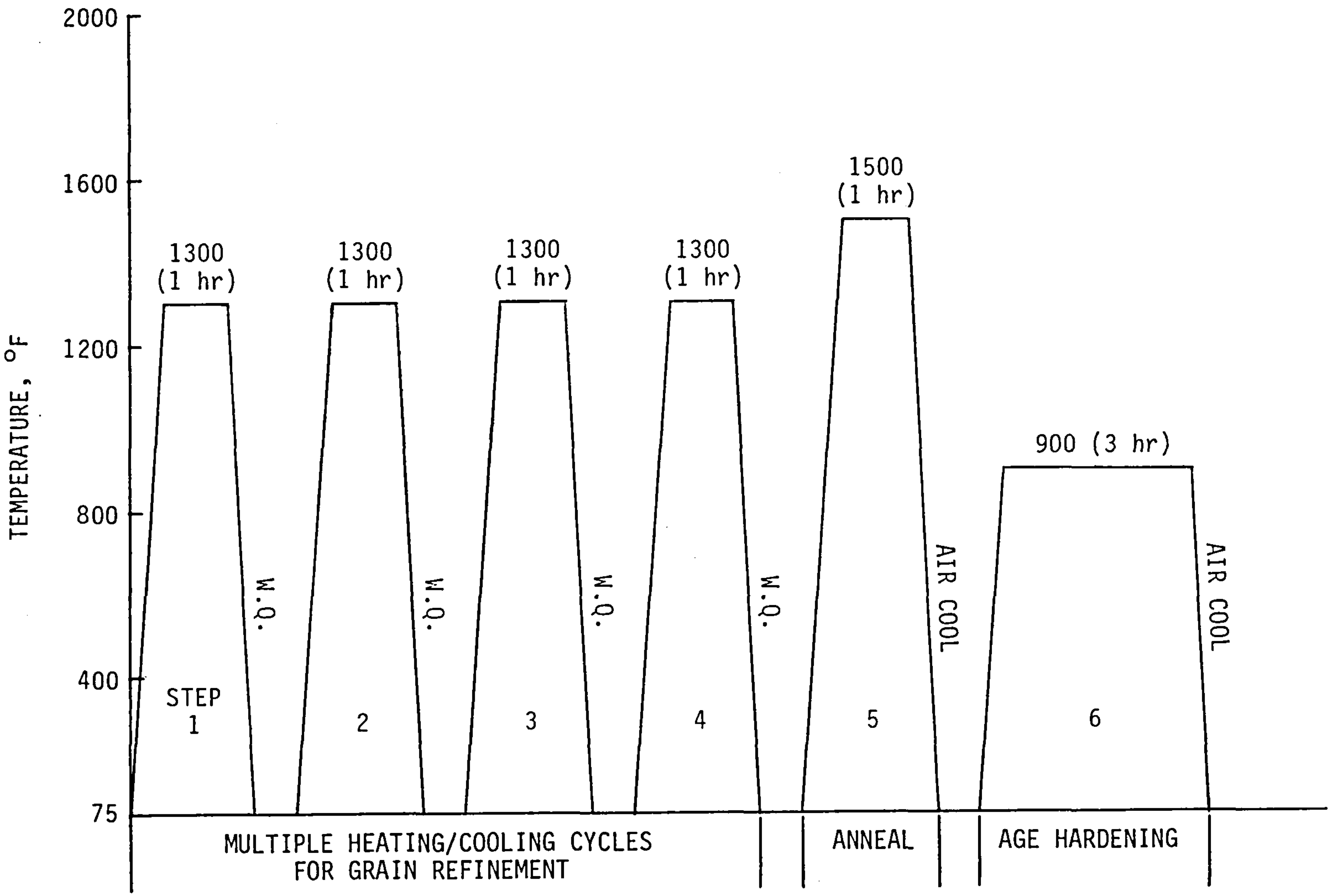


Figure 23.- Grain-refining heat treatment GR-1V for 18Ni 200 grade.

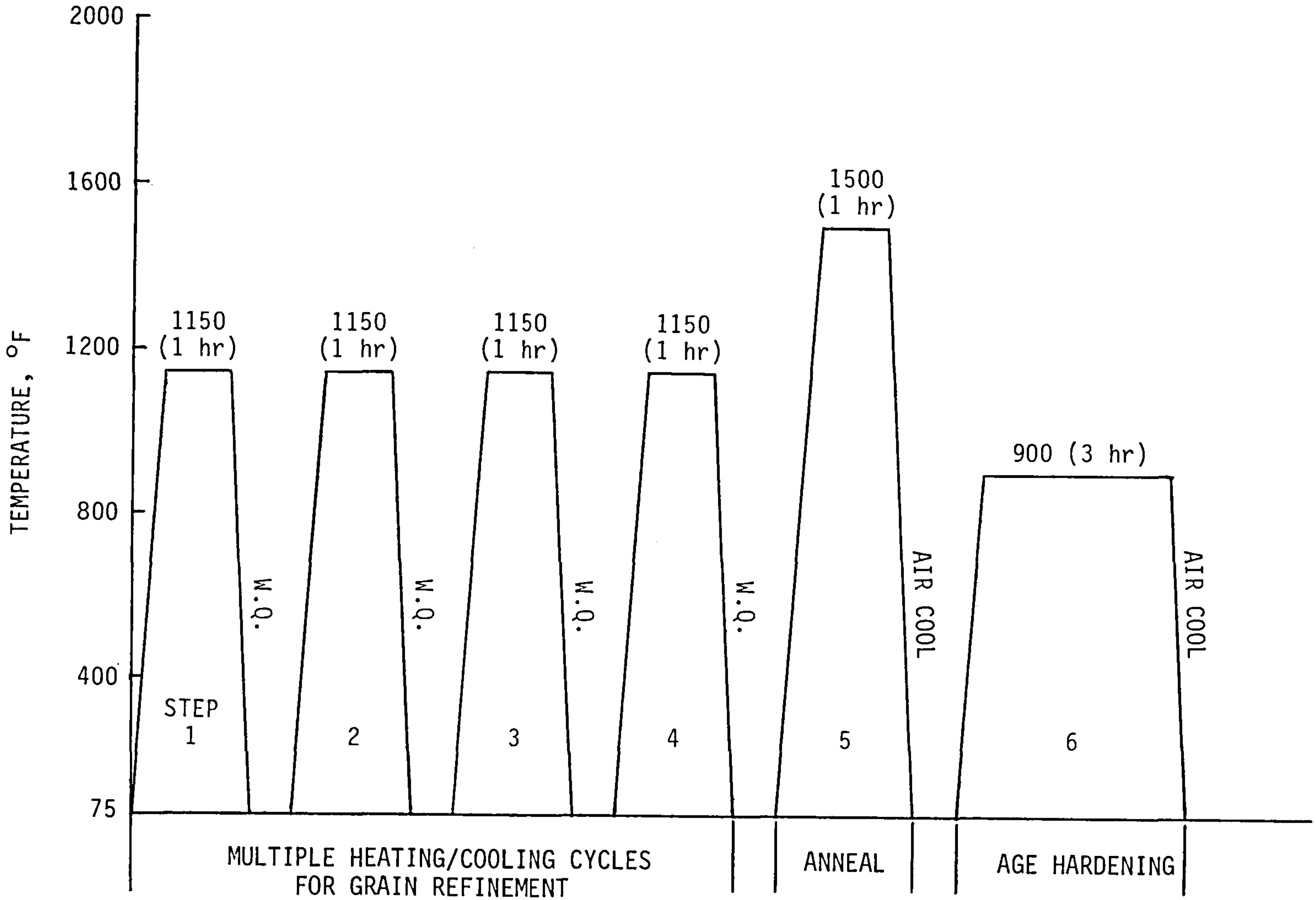


Figure 24.- Grain-refining heat treatment GR-2V for 18Ni 200 grade.

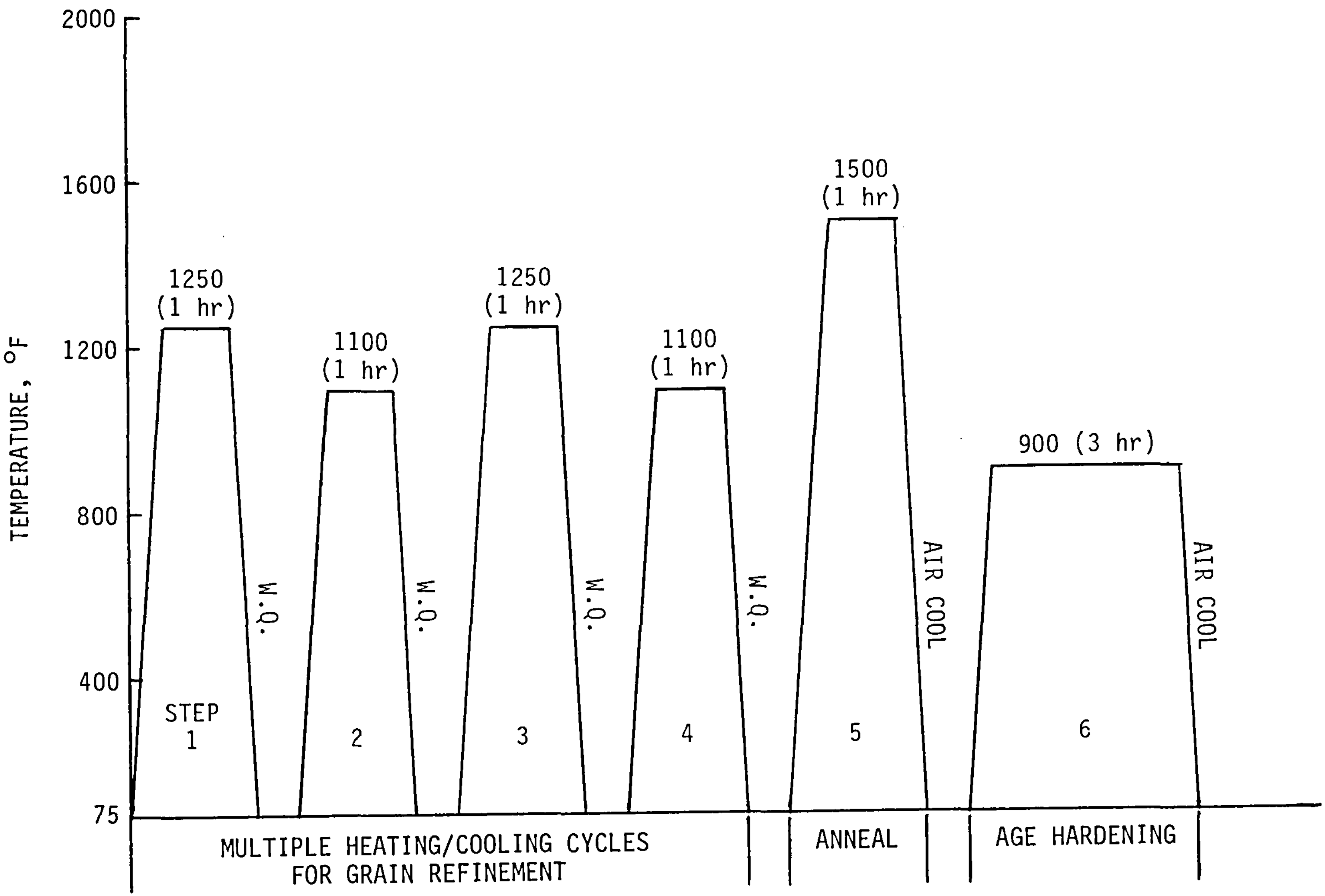
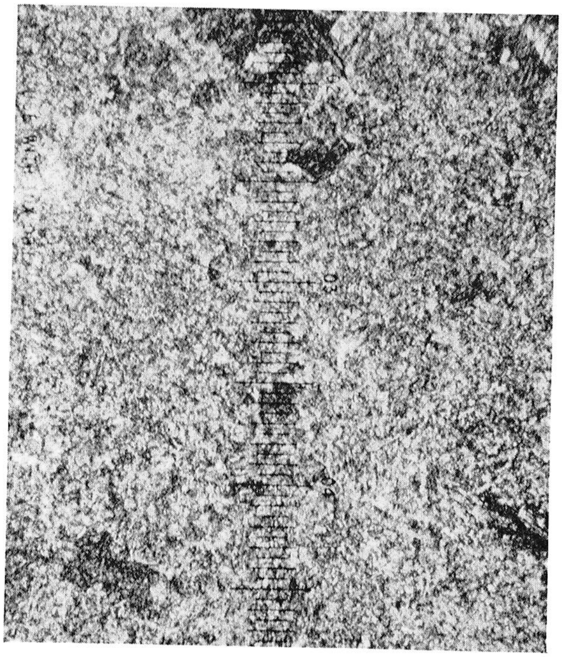
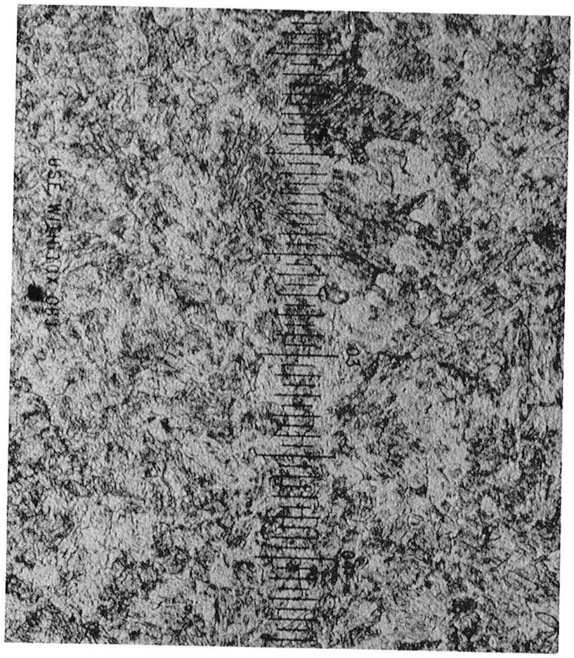


Figure 25.- Grain-refining heat treatment GR-3V for 18Ni 200 grade.



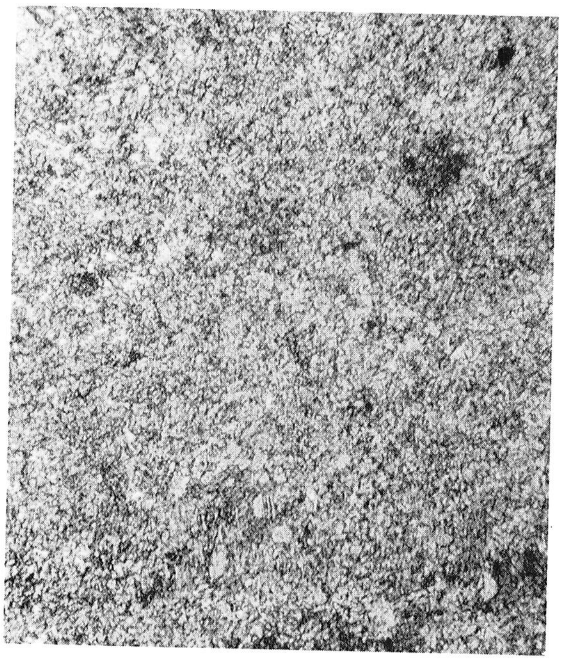
100 μm

Annealed, × 100



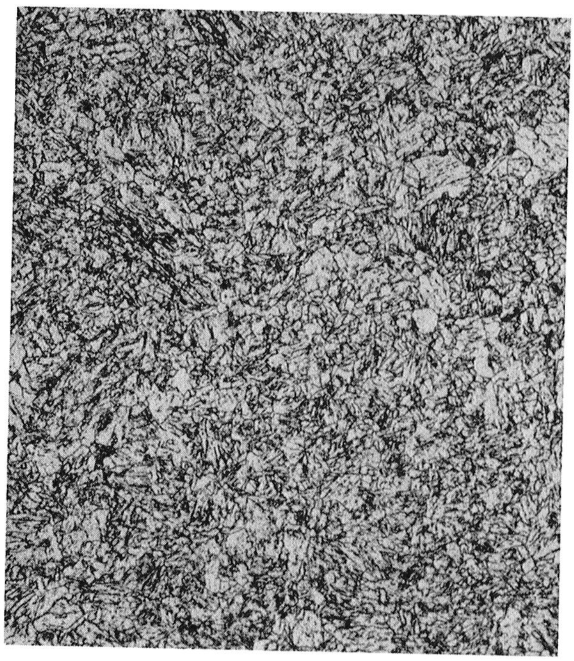
10 μm

Annealed, × 400



100 μm

Aged at 900°F, × 100



10 μm

Aged at 900°F, × 400

Figure 26.- Microstructure of 18Ni 200 grade GR-3V specimens.

L-84-90

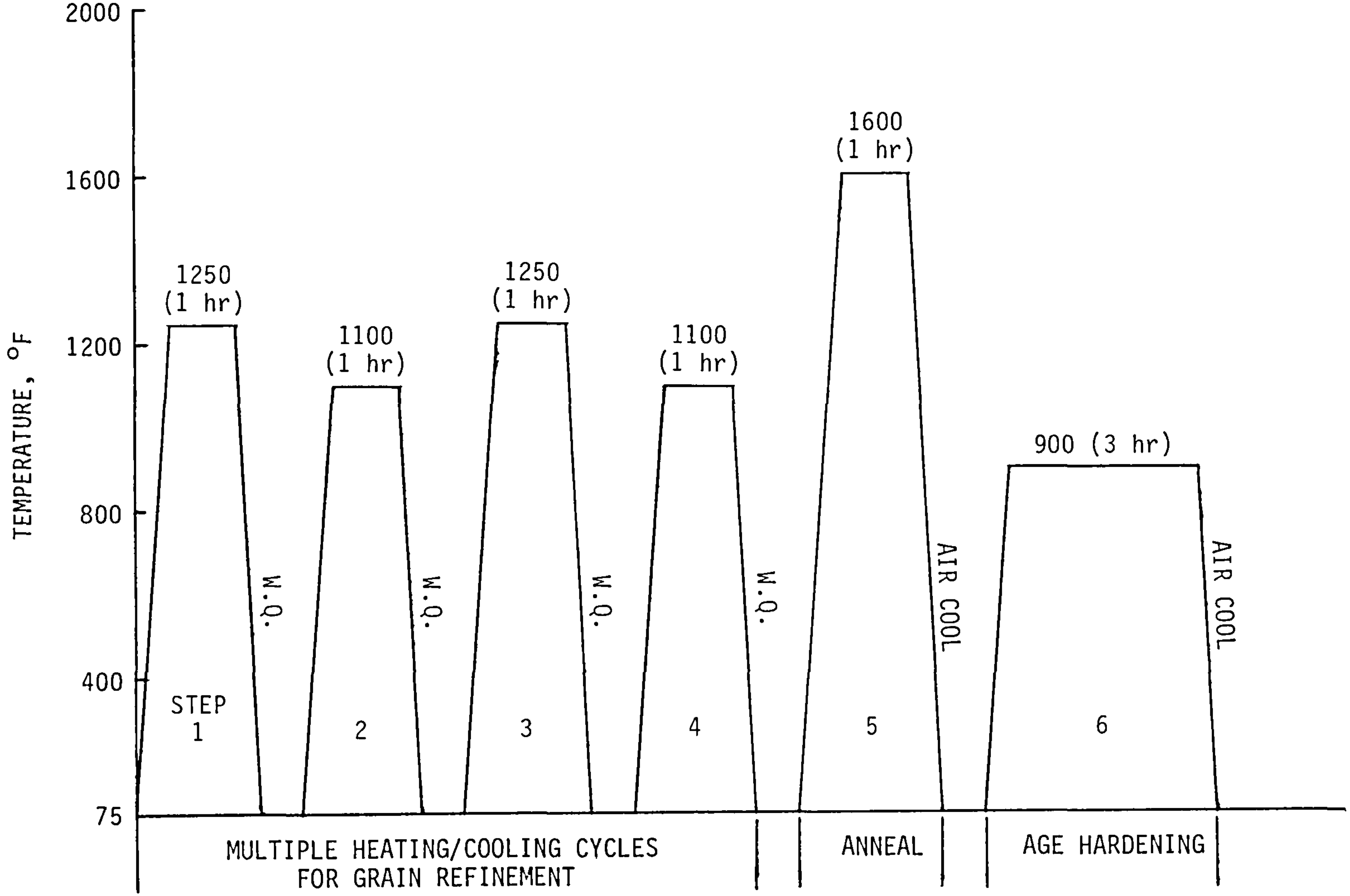
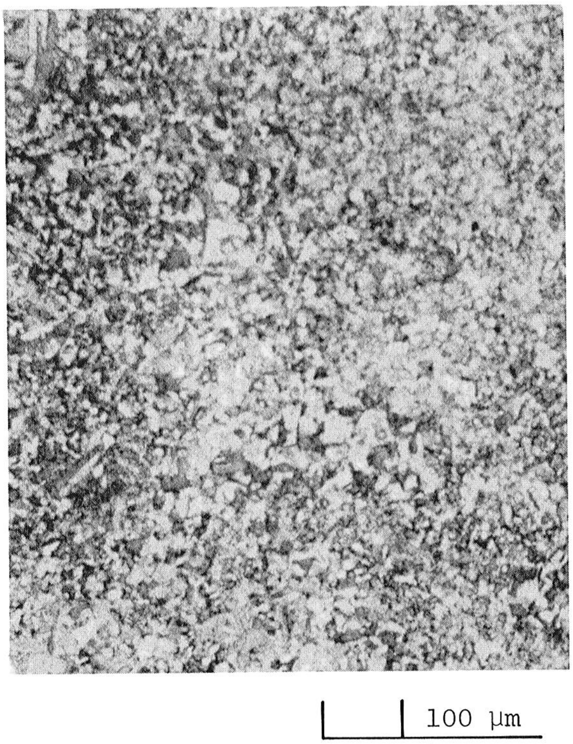
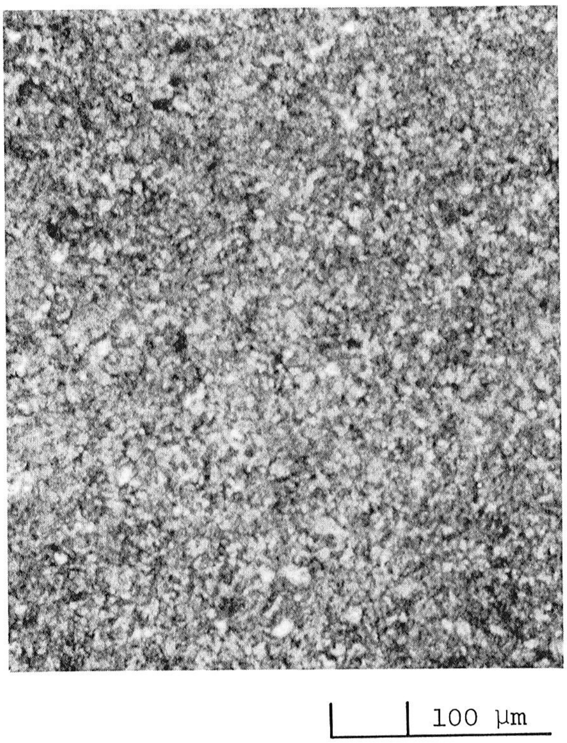


Figure 27.- Grain-refining heat treatment GR-4V for 18Ni 200 grade.



Annealed, × 100

Annealed, × 400

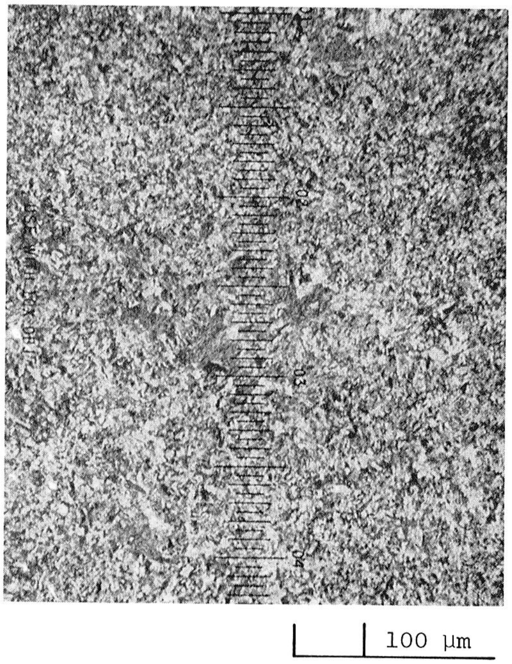


Aged at 900 °F, × 100

Aged at 900 °F, × 400

L-84-91

Figure 28.- Microstructure of 18Ni 200 grade GR-4V specimens.

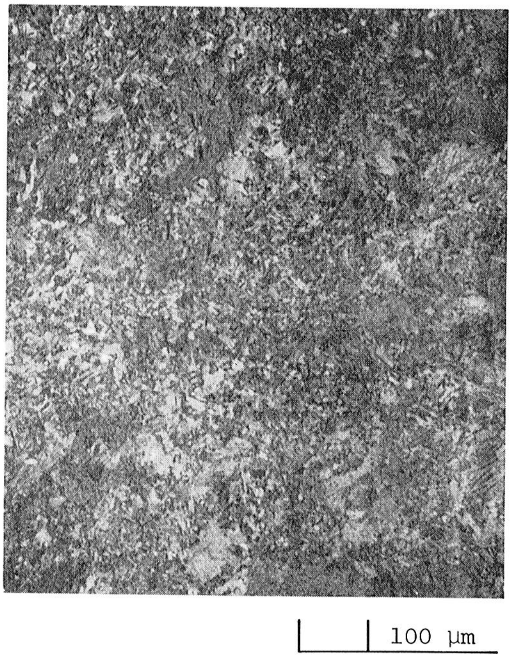


Aged at 1000°F, × 100

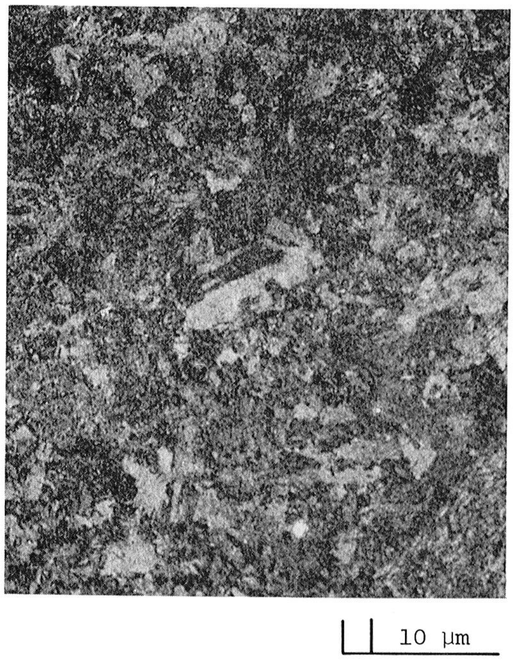
Aged at 1000°F, × 400

L-84-92

Figure 29.- Microstructure of 18Ni 200 grade GR-3V-1000 specimen.



Aged at 1100°F, × 100



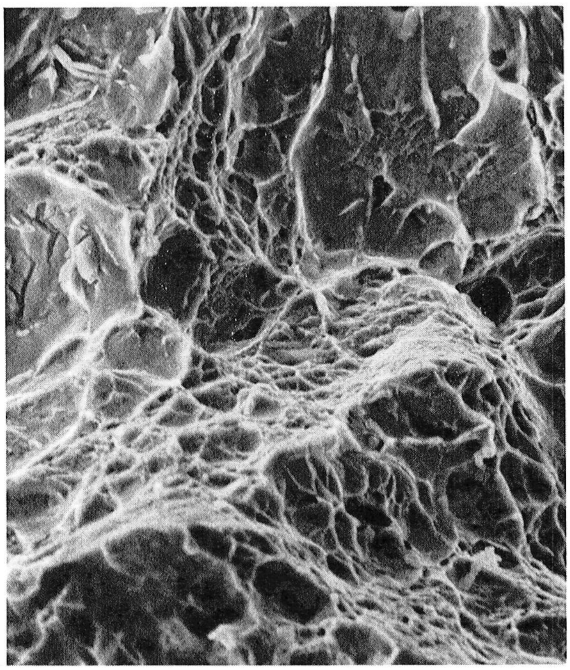
Aged at 1100°F, × 400

L-84-93

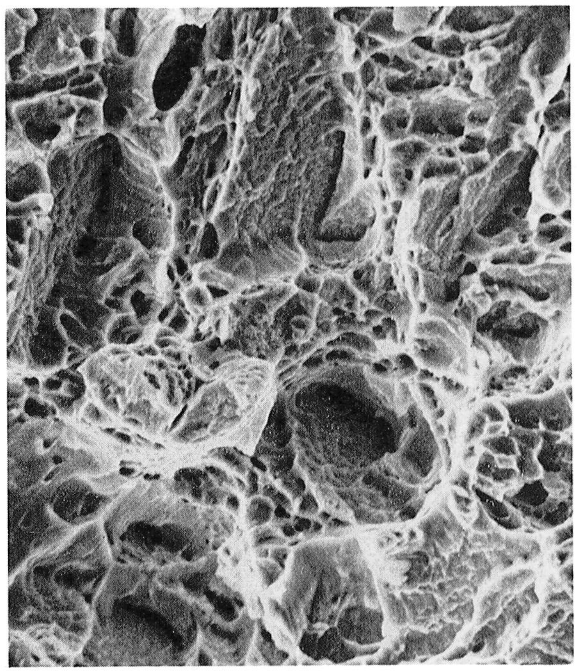
Figure 30.- Microstructure of 18Ni 200 grade GR-3V-1100 specimen.

GR-3V, × 1400

GR-4V, × 1400



GR-3V-1000, × 1400



Control, × 1400

L-84-94

Figure 31.- Fracture surfaces from Charpy V-notch test of 18Ni 200 grade specimens tested at -320°F.

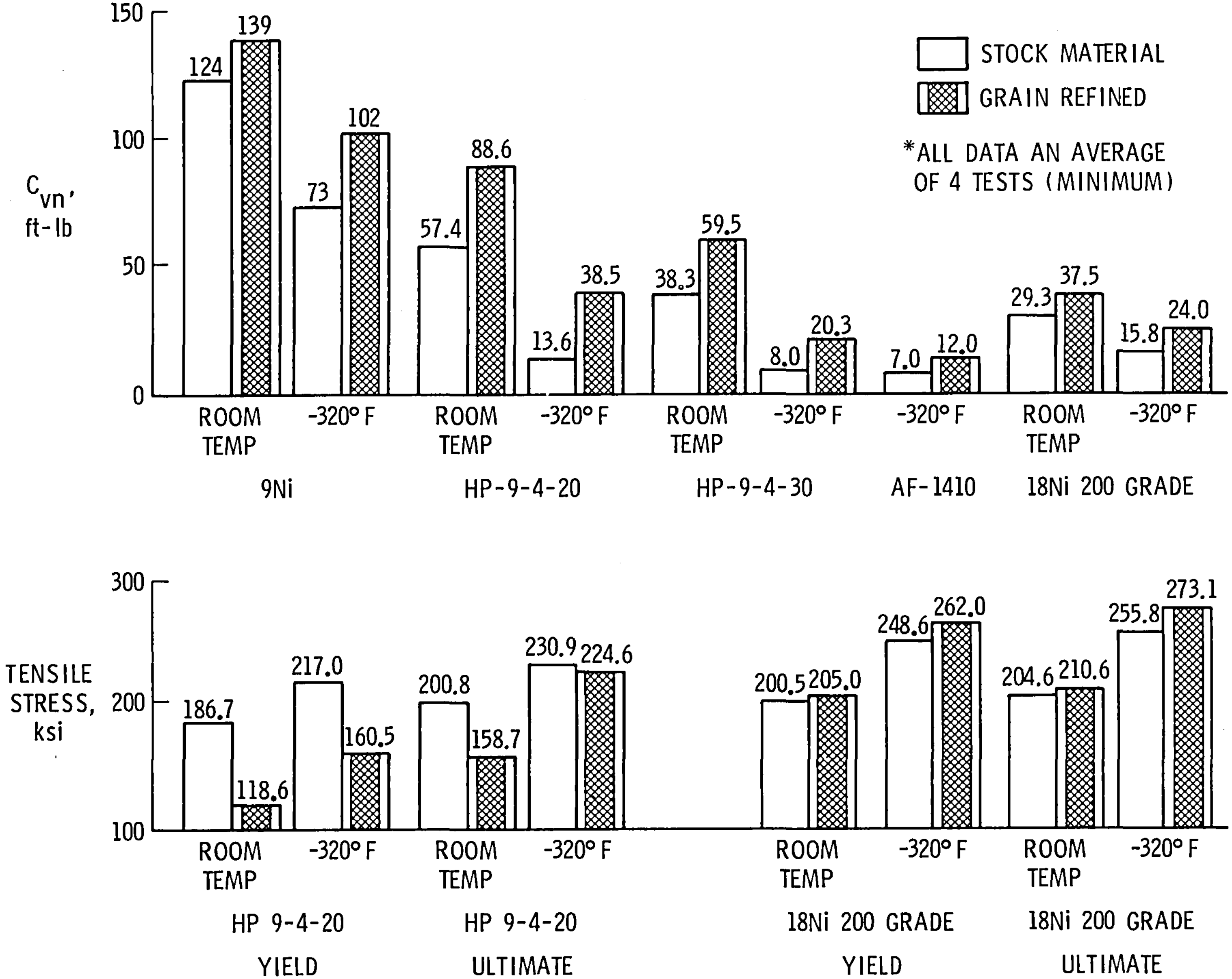


Figure 32.- Effect of grain refinement on mechanical properties.

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16. Abstract

The development of two high-Reynolds-number wind tunnels at NASA Langley Research Center which operate at cryogenic temperatures with high dynamic pressures has imposed severe requirements on materials for model construction. Existing commercial high-strength steels lack sufficient toughness to permit their safe use at temperatures approaching that of liquid nitrogen (-320°F). Therefore, a program to improve the cryogenic toughness of commercial high-strength steels was conducted. Significant improvement in the cryogenic toughness of commercial high-strength martensitic and maraging steels has been demonstrated through the use of grain-refining heat treatments. Charpy impact strength at -320°F was increased by 50 to 180 percent for the various alloys without significant loss in tensile strength. The grain sizes of the 9-percent Ni-Co alloys and 200-grade maraging steels were reduced to 1/10 of the original size or smaller, with the added benefit of improved machinability. This grain-refining technique should permit these alloys with ultimate strengths of 220 to 270 ksi to receive consideration for cryogenic service.

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