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ABSTRACT

Relationships between transverse tensile ductility and rolling and annealing schedules were investigated for solid-state sintered and annealed 80%W-8%Ni-2%Fe heavy alloy. Materials for this study were rolled at 900°C or 1150°C with varying reductions between anneals at either 1150°C or 1400°C. Final anneals of 1400°C and 1455°C and a 1000°C solution heat treatment were employed prior to tensile testing. Metallographic and fractographic analyses were performed to determine relationships between microstructure and physical properties.

Multiple 1400°C intermediate anneals with a maximum 60% rolling reduction produced higher transverse tensile elongations than were produced in materials that were rolled with a higher final reduction, e.g. 86%. Tensile elongation differences were attributed to the recrystallized intra-particle W grain sizes achieved during the final anneal. Materials that were given a maximum of 60% reduction prior to the final anneal had fewer intra-particle W grains and therefore higher ductilities; these effects are analogous to contiguity effects in liquid-phase sintered heavy alloys.

For materials rolled at either 900°C or 1150°C, no differences in transverse tensile elongation were observed. Materials that received 1150°C intermediate anneals had consistently lower ductility. Rolling at 900°C produced slightly higher elongations than rolling at 1150°C, but only when the material was annealed at 1455°C. Tensile yield and ultimate strengths did not vary greatly with the rolling and intermediate annealing conditions used during this study.

The occurrence of edge cracking correlated with the observed lateral spread and the material softness.

INTRODUCTION

Tungsten heavy alloys have found application in radiation shielding, for which alloys of 70% to 90% W have been produced commercially in sheet form. Additional applications for these alloys are possible if they can be produced with adequate ductility. Ductility improvement obtained by reducing the W content in favor of a higher content of ductile matrix comes with the cost of lowered density (1). An alloy of 80% W represents a compromise between density and ductility. In addition, the alloy is solid-state sintered to a rollable, uniform thickness billet, which is not possible by liquid-phase (LP) sintering, and sheet rolled from the alloy can be solid-state annealed to high ductility, 30 to 35% tensile elongation. However, sheet produced from these alloys has exhibited variable ductility in the as-annealed state, even though none of the common embrittling mechanisms, such as hydrogen and impurity embrittlement (2), have been found. Similar variability has been observed for 90% W alloy but not in the 70% W alloy.

Yodogawa (3) showed that the recrystallization of the W phase particles in 90% W alloy during annealing severely diminished longitudinal ductility. After LP-sintered 90% W is cold rolled to reductions of the greater than 70%, 1-h anneals had little effect on ductility until the annealing temperature reached 1000°C, at which point the ductility dropped precipitously due to recrystallization of the W. On the other hand, after 30% cold rolling reduction, ductility continued to increase with annealing temperature, reaching a maximum of 17% elongation at the highest annealing temperature, 1420°C; very little recrystallization occurred at any annealing temperature after rolling 30%. Presumably, the detrimental effect of recrystallization is due to the increased amount of weak W-W grain boundaries within the W (2,4).

The object of this study was to develop rolling parameters that maximize ductility in the 80% W alloy by minimizing the development of weak W-W grain boundaries alloy during rolling and annealing. The parameters thought most important were investigated: rolling temperature (1150°C and 900°C), intermediate annealing temperature (1400°C and 1150°C), and rolling reduction between anneals (27% to 86%). Since transverse tensile properties, especially elongation, are more sensitively affected by processing variations than are longitudinal properties, characterization was performed using transverse tensile testing, optical metallography, and scanning electron fractography were performed on selected tensile specimens.

EXPERIMENTAL

The material used for this study was supplied by Teledyne Firth Sterling and was in the form of an as-sintered slab nominally 8.5 in. wide by 18 in. long by 1-1/8 in. thick. The composition was nominally the 80%W-8%Ni-2%Fe alloy solid-state-sintered to near 100% of the 15.4 g/cm^3 theoretical density.

Rolling Trials

The as-received slab was sectioned equally into six rolling billets, 4 in. by 6 in. by 1-1/8 in. thick. Preliminary rolling trials on as-sintered billets showed that 1) rolling at 1200°C preheat temperature allowed reductions of >75%, and at 1000°C billets split with reductions of >50%, 2) the billet had to be reheated every pass to prevent splitting, 3) 10% to 15% reductions per pass were satisfactory, whereas 20% caused early splitting, and 4) rolling speed variations of 30 to 75 surface feet per minute (sfpm) were inconsequential. To improve billet density and thickness uniformity, breakdown rolling of the six as-sintered billets was done at 1150°C, in two 5% reductions at 40 sfpm, followed by a "homogenization" anneal at 1400°C for 3 h in 50%Ar-50%H_2. These six billets were then cut into twelve, half-length billets.

In subsequent steps the rolling schedule was varied for each billet. Preheating for rolling was done in air at either 900°C or 1150°C. Annealing was done in 50%Ar-50%H_2 either at 1150°C for 16 h or at 1400°C for 3 h. Reductions between anneals were either increased in the progression 27%, 38%, 45%, and 60%, or abbreviated to 27% and 86%. A single additional billet was rolled with the same final reduction used in the progressive schedule, 60%, preceded by a 75% reduction. All three schedules yielded a final thickness of 0.1 inch. Figure 1 schematically describes the three reduction schedules used in terms of thickness reduction and the number of intermediate anneals.

Rolling for this study was performed using 14-in.-dia. by 18-in.-wide rolls installed in 900 kip separating force Waterbury Farrell Rolling Mill.

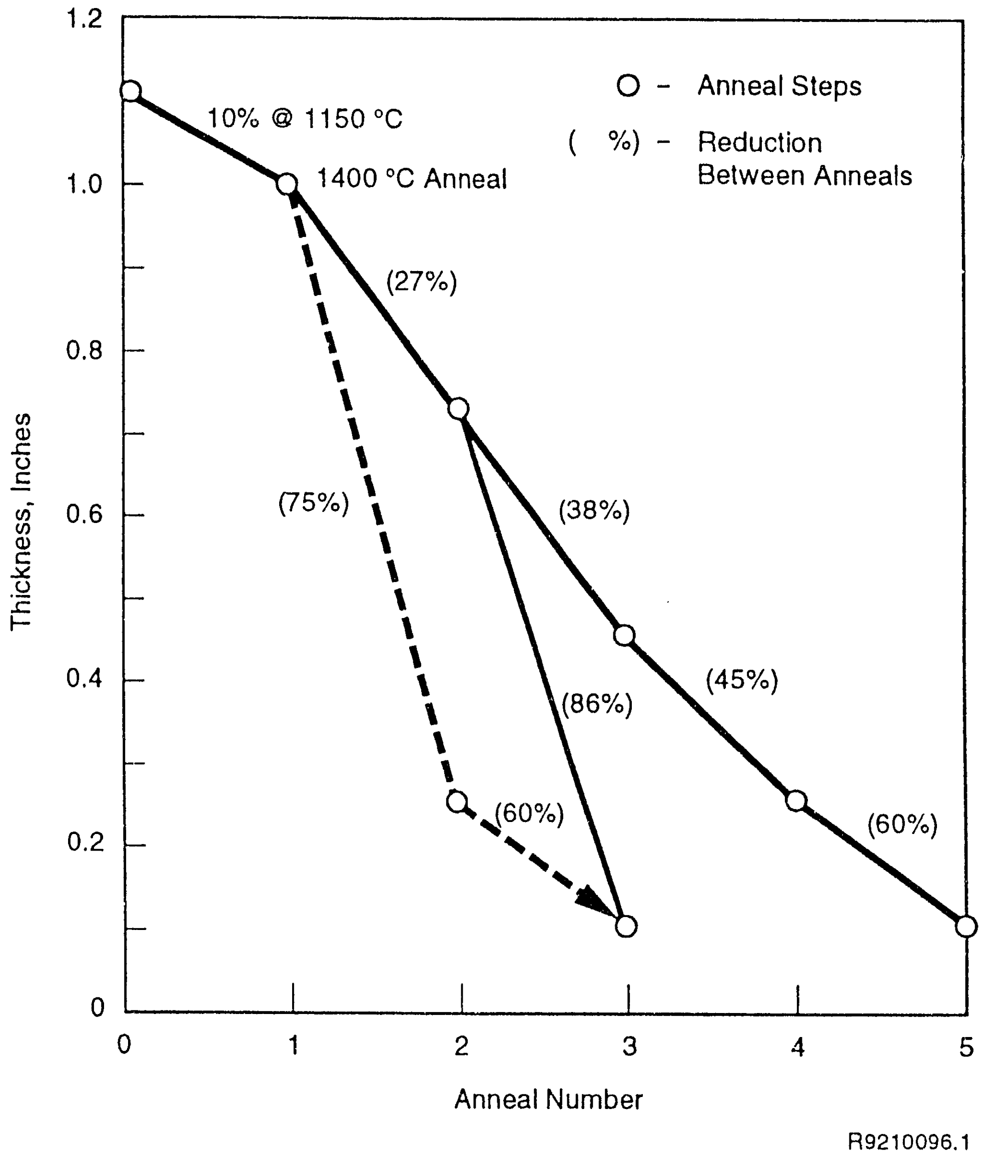


FIGURE 1. Rolling Schedules used for 80% W Heavy Alloy Slabs

With the exception of the first two passes, the billets were reduced 7.5 to 10% per pass and the number of passes varied from a minimum of 28 to maximum of 32. The first two passes were performed at 5% as explained earlier in this section. The billets were rolled without cross rolling, and the ends were reversed between each pass.

The billets were reheated between each pass in air using the minimum time required to reheat the billet. Minimum reheat times were used to minimize the annealing effects of the 1150°C rolling temperature. A second order effect of short preheat times is the minimization of oxidation, which is significant particularly with the 1150°C rolling temperatures. Times for preheat were 13 min for the 1.12-in.-thick billet. Reheat times started at 8 min for 1 in. and greater thickness and decreased with decreasing thickness to a minimum of 1 min at thicknesses less than 0.150 in.

The billets were rolled at 40 sfpm until 0.2 in. thickness was achieved. The speed was then increased to 55 sfpm until the thickness reached 0.15 in., at which time the mill speed was increased to 75 sfpm. The mill speed was increased with decreasing thickness in order to minimize quenching effects of the rolls, thereby improving the temperature uniformity of the experiment and decreasing the reheat time between passes.

Final Heat treatment and Tensile Testing

Final annealing treatments of either 1400°C for 4 h or 1455°C for 16 h in 96%Ar-4%H_2 were employed using 2-h ramps to temperature followed by rapid cooling from the annealing temperature. After final annealing all sheets were given a vacuum outgassing and solution heat treatment of 1000°C for 1 h followed by a water quench. The objective of this heat treatment was to prevent degradation of ductility due to impurity segregation, and to a lessor extent, hydrogen embrittlement.

Quasi-Static tensile testing was performed using flat tensile specimens with a gage dimensions of 0.25 in. by 0.1 in. (as-rolled thickness) by 1 in. long. Specimens were machined from fully heat treated sheet blanks in the transverse orientation and tested at a strain rate of 7×10^{-4} in./in.-s. Specimens were taken toward the mid-length of the sheets to avoid material from near the as-sintered slab edge; no material within a slab thickness, 1 1/8 inch, of the slab edge was sampled for tensile testing. Three consistent specimens were tested for each rolling schedule and final anneal.

EXPERIMENTAL RESULTS AND DISCUSSIONS

Tensile strengths for all specimens tested were 84 to 96 ksi yield and 140 to 150 ksi ultimate, the higher values resulting from the 1400°C anneal and the lower values resulting from the 1455°C anneal; these differences are small. However, highly significant differences in elongation were obtained for the various rolling schedules, as shown in Table 1.

Annealed samples rolled with a final reduction of 60% consistently had elongations which were higher than samples receiving 86% final reductions. It is especially noteworthy that (for sheets rolled at 900°C with just two intermediate anneals at 1400°C) the 75% to 60% reduction schedule produced optimal tensile elongations of 32%, whereas the 27% to 86% schedule yielded only 22% elongation.

TABLE 1. Annealed Transverse Tensile Elongation

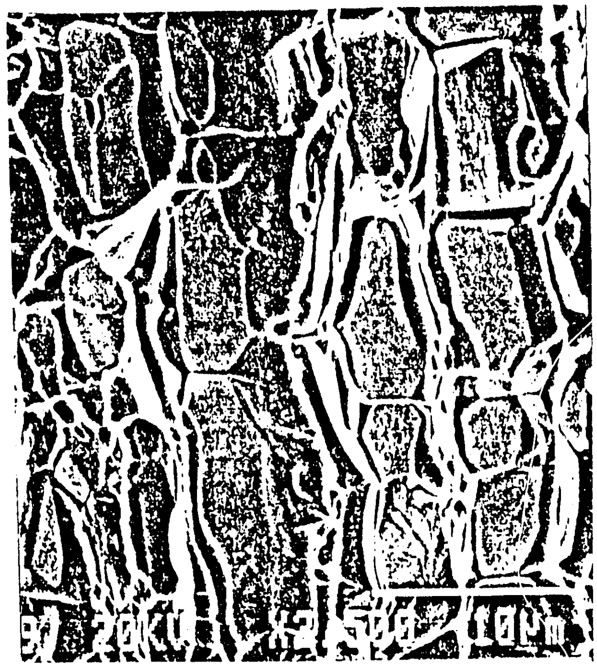
	Transverse Tensile Elongation, %			
	1150°C Rolling Temperature		900°C Rolling Temperature	
	1400°C-3 h Intermediate Anneals	1150°C-16 h Intermediate Anneals	1400°C-3 h Intermediate Anneals	1150°C-16 h Intermediate Anneals
80% Final Reduction				
4 intermediate anneals	32 (31)[(a)]	27 (26.5)	32 (33)	27 (29)
2 intermediate anneals			32	
86% Final Reduction				
2 intermediate anneals	23 (25)	20 (23)	22 (28)	19 (25)

(a) Values are for elongation after final anneals at 1400°C for 4 h. Values for elongation after 1455°C-16 h anneals are given in parentheses.

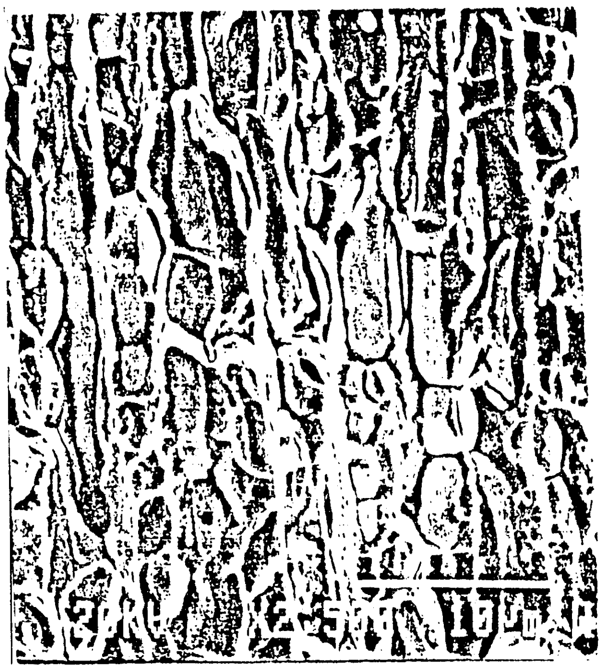
The effect of rolling reduction on ductility was also apparent in the fractography. As shown by Figure 2b, the samples that received 86% reductions had a predominance of W intergranular failure at recrystallized grain boundaries. In contrast, samples that received 60% final reduction (Figure 2a) had a predominance of cleavage fracture through the particles and ductile rupture of the matrix. Consistent with previous studies (2-4), cleavage of W particles and ductile rupture of matrix in W heavy alloy is an indication of high ductility; W-W interfacial failure at recrystallized grain boundaries is analogous to W-W interfacial failure at contiguous W particles, which dominates the behavior of very high W content alloys (e.g., >93% W). W-W interfaces are the weakest link in the heavy alloy structure and they always appear in the fracture surface when they are near the fracture path. A predominance of W-W interfacial or intergranular failure always indicates embrittlement.

In an attempt to anneal the W particles to single crystals, an extremely high temperature (1455°C) anneal for 16 h was tried. This annealing temperature is within 5°C of the melting point of the matrix, and would be very difficult to reproduce in a production furnace; however, it does indicate the ultimate potential of solid-state annealing. As shown in Table 1, the 1455°C for 16 h final anneal partially recovered elongation in the low-ductility, high-final-reduction materials. However, the increase in ductility did not correspond to a change in fracture mechanism, as shown by the SEM fractograph in Figure 2c, which is directly comparable to the sample annealed at 1400°C shown in Figure 2b. Fracture occurred by W intergranular separation and ductile rupture of the matrix. The effect of the final anneal on the microstructure is shown by comparing Figure 3b and 3c. Notice that the W particle has become much larger and more rounded by the extreme anneal at 1455°C; however the W particles are still polycrystalline.

(a) 60% Final Reduction 1400°C-4 h Final Anneal 32% Elongation

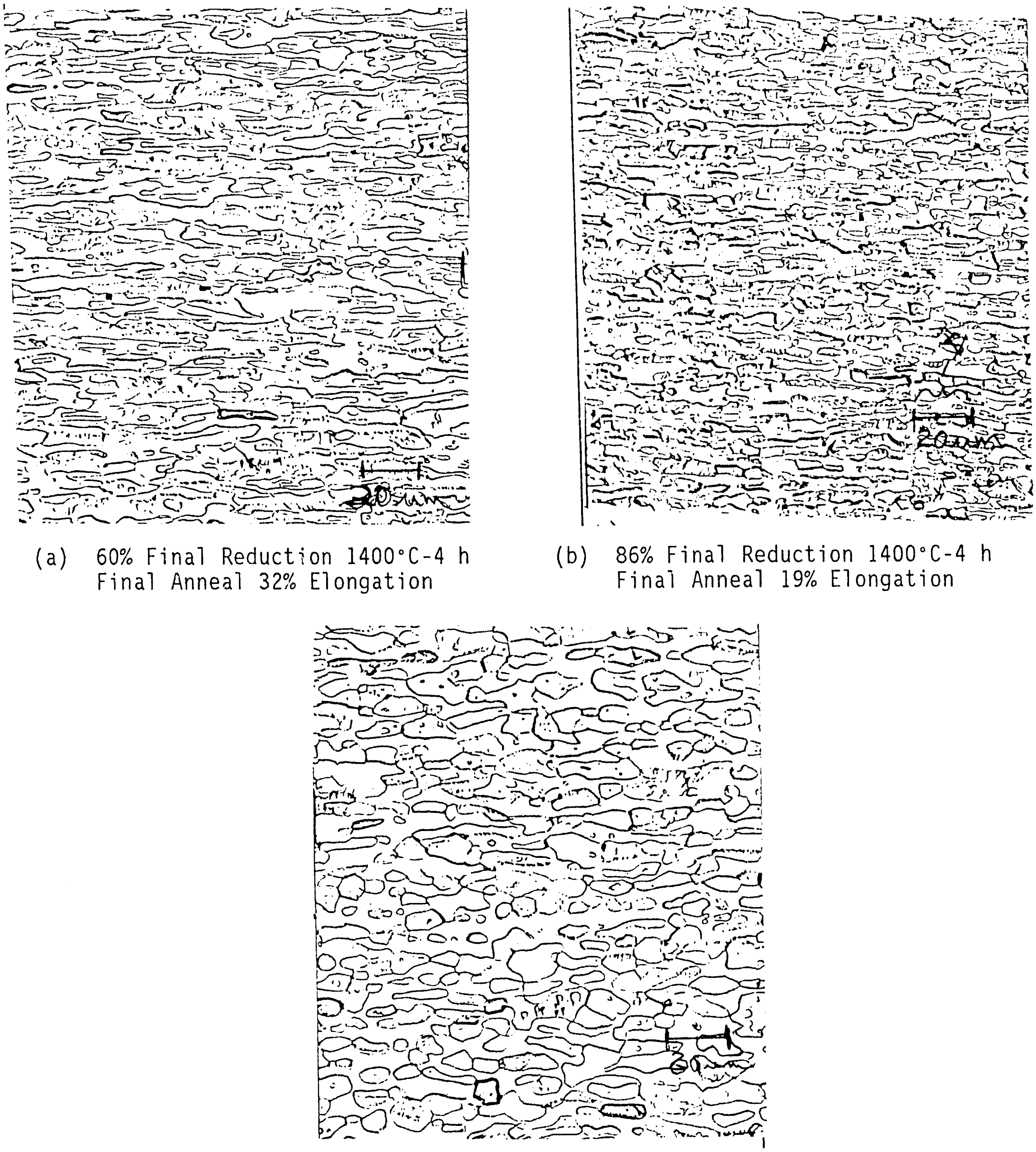


(b) 86% Final Reduction 1400°C-4 h Final Anneal 19% Elongation



(c) 86% Final Reduction 1400°C-16 h Final Anneal 25% Elongation

FIGURE 2. SEM Fractographs of Tensile Specimens. Rolling direction was up-down on page. Tensile direction normal to page. Highest elongation, a, has predominately W cleavage. Lowest elongation, b, has predominately W-W interfacial failure. Same material as b but with extreme anneal, c, exhibits W particle growth. W-W interfacial failure still predominates. Corresponding microstructures shown in Figure 3.



(a) 60% Final Reduction 1400°C-4 h
Final Anneal 32% Elongation

(b) 86% Final Reduction 1400°C-4 h
Final Anneal 19% Elongation

(c) 86% Final Reduction 1400°C-16 h
Final Anneal 25% Elongation

FIGURE 3. Optical Microstructures of Tensile Specimens. Rolling direction was up-down on page. Tensile direction normal to page. Highest elongation, a, has predominately single crystal W particles. Lowest elongation, b, has many more multicrystal W particles. Same material as b but with extreme anneal, c, has coarsened W particles and less W-W interfacial area than b. Elongation is still less than a. Corresponding fractographs shown in Figure 2.

As shown by the optical metallography of Figure 3a, the final microstructures for materials that had elongations of 32% had few intra-W grain boundaries. Conversely, materials that had elongation of 19% (shown in Figure 3b) had polycrystalline W-particles. The resulting microstructures were a function of final reduction and, to a lesser extent, intermediate annealing temperature. As shown by transverse tensile elongation data given in Table 1, materials rolled using the 1150°C intermediate had consistently lower elongation than materials rolled using 1400°C intermediate anneals. The resulting polycrystalline W-particle microstructures, and corresponding low transverse elongation, for materials rolled using 1150°C intermediate anneals, suggest that complete recovery has not occurred and that upon final anneal there was nucleation of multiple grains.

These results have implications for other W content heavy alloys. Higher W content alloys should be more sensitive to rolling schedule than lower W content alloys. Yodogawa (3) demonstrated the marked detrimental effect of high rolling reductions on W recrystallization and ductility in the 90% W alloy. Conversely, the 70% W alloy has shown only minor variability in ductility, both in laboratory and commercial processing, which implies that the 70% W alloy is relatively insensitive to rolling schedule.

Table 1 also indicates that variations in ductility obtained from the two rolling temperatures is negligibly small when the rest of the rolling and annealing schedule was identical. Therefore, it appears that deformation of the 80% W heavy alloy at 900°C and 1150°C results in a microstructure that has a similar amount of residual "cold work" and that the annealing response, at both 1400° and 1455°C, for the two rolling temperatures was the same.

Although the annealing responses of the materials rolled at 900°C and 1500°C were the same, the behavior during rolling was different. The loads required for deformation were approximately 20% higher for the 900°C rolling temperature. Also, materials rolled at 900°C exhibited less edge cracking than did materials rolled at 1150°C although the total edge spread of the samples was the same for a given reduction and annealing schedule. Table 2 provides a summary of the edge spread and edge cracking measurements for the eight rolling schedules. The thickness at which edge cracking started is also given in Table 2. The edges of the "softest" sheets cracked the most and the earliest in the reduction; the softest sheets were those rolled at 1150°C and given multiple 1400°C intermediate anneals. Figure 4 shows the cracking for selected

TABLE 2. Edge Cracking Related to Rolling and Annealing Temperatures and Final Reduction

Rolling Temperature	1150°C				900°C			
Intermediate Anneal Temperature	1400°C-3 h		1150°C-16 h		1400°C-3 h		1150°C-16 h	
Final Reduction	60%	86%	60%	86%	60%	86%	60%	86%
Lateral Spread, %	4	13	4.5	4.5	4	10	4	6
Edge Crack Depth, in.	0.125	0.375	None	0.25	None	0.313	None	None
Crack Start Thickness, in.	0.151	0.345	N/A	0.226	N/A	0.235	N/A	N/A

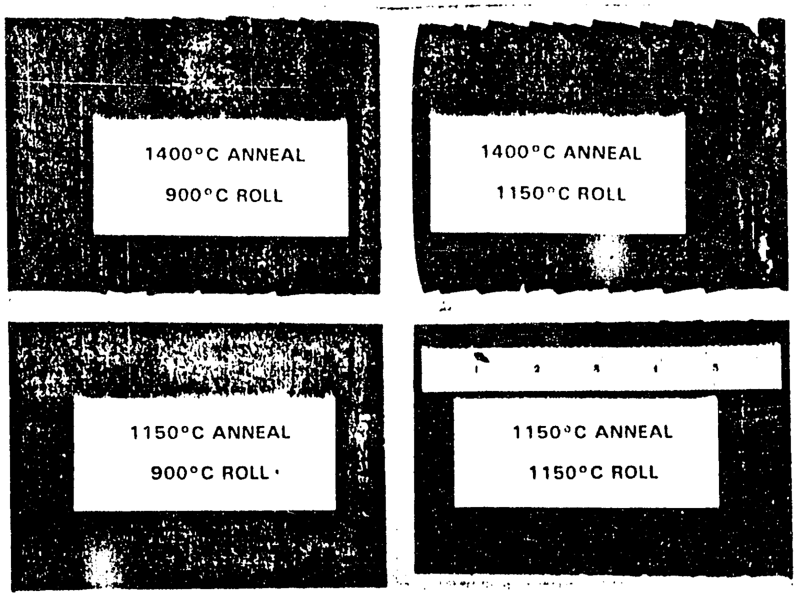


FIGURE 4. Extremes of Edge Cracking Behavior as Affected by Rolling and Intermediate Annealing Temperatures. All four sheets pictured had a final reduction of 86%.

sheets rolled 86% in the final reduction; these four sheets show the range of cracking behavior experienced for the four combinations of intermediate annealing and rolling temperatures. The sheets rolled and intermediate-annealed at the lower temperatures exhibit little or no edge cracking when compared with materials processed at the higher temperatures.

In no case did the edge cracking lead to splitting of the sheet rolled according to the schedules used in this study; in fact, if the edges of the sheet are to be sheared to width after rolling, edge cracking is of no practical consequence. Splitting observed in previous studies appears to be related more to billet defects, low rolling temperatures, and excessive reductions between anneals.

CONCLUSIONS

1. Recrystallization behavior of the W particles is critical to the ductility of the 80% W heavy alloy; formation of polycrystal W particles is detrimental to ductility.

2. The W particle recrystallization behavior was strongly a function of the maximum rolling reduction before the final anneal. Sheets that were rolled 60% before final annealing had higher elongations and fewer intra-particle W grains than did samples rolled 86%.

3. Consistently higher elongations were produced with intermediate anneals at 1400°C than at 1150°C. Presumably, at 1400°C more complete recovery occurs within the W, which diminishes the driving force for recrystallization of multiple grains. The elimination of new W grain boundaries by grain growth is extremely sluggish and cannot be accomplished without W particle growth.

4. The depth of edge cracking is related to material softness and the resulting lateral spreading of the sheet. With proper rolling schedules, edge cracking does not lead directly to splitting of the sheet and is inconsequential.

RECOMMENDATIONS

The fabrication of ductile W heavy alloys with circa 80% W provides a difficulty in the control of the W-particle microstructure. However, there are some technically feasible options:

1. Solid-state-single crystal growth methods such as strain-and-anneal and directional, higher thermal gradient anneals look promising for manipulation of W particle structures.

2. Liquid-phase resinter techniques such as high-temperature-gradient zone melting may develop single crystal W particles without undue segregation (Gurwell 1985).

Other, conventional options are available for higher and lower W content alloys and should be used where possible. For instance, alloys of 90% W and greater (and possibly as low as 85% W) can be liquid-phase resintered to high ductility (and single crystal W particles) with conventional furnacing techniques. Also, 70% W alloy has been routinely annealed in the solid state to high ductilities, probably because the very high volume fraction of matrix, 56%, dominates the behavior of the alloy; there is also very little contiguity of W grains observed in the 70% W microstructures.

ACKNOWLEDGEMENTS

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