JMEPEG (1998) 7:733-738

AR-Aging as a New Approach for Enhanced Results

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(Submitted 16 December 1997; in revised form 25 August 1998)

MAR-aging steels have earned a niche in the metal market arena, especially where aerospace and outer-space applications are concerned. MAR-aging steels owe their high strength, excellent fracture toughness, and good ductility to a precipitation-hardening (aging) mechanism that has been debated by scientists for several years. Because of today's trend toward more demanding design requirements and a continuing need to better understand the MAR-aging family of materials, six different alloys (C-200, C-250, C-300, C-350, T-250, and T-300) were selected for study using a singular processing treatment: a hot-wall zone-gradient furnace. These alloys were evaluated for the effects of a specific thermal gradient (°C/cm) from 1231 °C (2250 °F) at the hot-wall limit to about 260 °C (500 °F) at the opposite end, the cold wall. All six alloys were evaluated in terms of their microstructure, microhardness, composition, and associated properties as a result of this specific thermal processing method. In this paper, detailed observations on the C-350 alloy are presented, and the results are interpreted in terms of a new heat treatment cycle called AR-aging.

Keywords isochronal heat treatment, MAR-aging steel alloy C-350, polygonization, recrystallization and recovery, zone gradient profile

1. History

Both discovery and development of the early MAR-aging alloys are attributed to the work (Ref 1-4) of C.G. Bieber, R.F. Decker, S. Floreen, and others under the sponsorship of the International Nickel Co., Inc. (INCO). Production melts of these alloys were made at Latrobe, Pennsylvania by Vasco. The original impetus for these new alloys was an improved hull for a nuclear submarine. Since then, MAR-aging steels have been successful in a multitude of other application areas.

Unfortunately, the specifics of MAR-aging metallurgy are not well understood by most engineers and technologists. This problem is a result of economic factors that have caused downsizing, mergers, and elimination of the research staffs that were assigned to this "minority market" of MAR-aging steels. It suffices to say that contributions to the sum of total knowledge for MAR-aging alloys have been inadequate relative to the needs and priorities of the end user. As a result, more field failures have occurred with MAR-aging steels than with other new alloys from the same era, particularly where air-melted material was involved. Consequently, this paper is dedicated to a better understanding of these special alloys.

2. Materials

All six alloys for this investigation were manufactured by the method of IVM-CVM using state-of-the-art melting practice. Ingots were reduced to round bar using a standard reduction sequence at the producer mill, Vasco. Nominal composition of the alloys that were studied in this investigation are given in Table 1. As shown in Table 1, these alloys represent both iron-cobalt types (C-grades) and iron-titanium types (T-grades).

Each round bar was about 305 mm (12 in.) long with an average diameter of 19.1 mm (0.75 in.). Exteriors of each round bar were ground smooth to eliminate surface oxides and seams from the fabrication cycle. Twelve bars (two of each alloy type) were thermally processed for this investigation.

3. Methods

Each load for the zone gradient furnace used four bars (two alloy types per run), two of which were instrumented with thermocouples along their length dimension and two that were not instrumented. The two instrumented bars were always placed on the outside of the other two bars, and similar alloy types were located next to each other. A Model GF-14 SATEC (SATEC Systems Inc., Grove City, PA) gradient furnace was used with a hot back-wall temperature of 1231 °C (2250 °F) for a one hour isochronal heat treatment cycle. Temperature (T) versus distance (D) from the hot-wall end was monitored and recorded for each furnace cycle.

Test bars were measured for length and diameter changes as a function of this thermal gradient processing. Both optical and electron metallography were applied along with Vickers microhardness testing (300 g load, 15 s dwell time) to characterize changes in structure that were caused by these thermal effects. About thirty specimens were obtained from each bar with parallel serial sectioning using a diamond saw wafering machine.

4. Discussion

Typical thermocouple data for a given run of temperature (T) versus distance (D) revealed a gradual curvilinear response from hot-wall to cold-wall that is indicative of recovery and recrystallization effects. Figure 1 shows how these observations varied for alloy C-350. In Fig. 1, the squares reveal the actual

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observations for this specific alloy with its associated best-fit, linear-correlation curve for alloy C-350 data.

Figure 2 shows the process of softening where the effects of a hypothetical isochronal annealing treatment are presented. If AR-aging is to be better understood, Fig. 2 is required for definition purposes.

Here, the Rockwell C hardness (HRC) decreases as the process temperature (*T*) increases with three important changes of state being observed for a given stored energy. These important process cycles of recovery, recrystallization, and grain growth have been described in detail by Margolin (Ref 5).

Figure 3 reveals the response for alloy C-350 after a one hour soak within the zone gradient SATEC furnace. In Fig. 3, Vickers microhardness (HV) undergoes a reversal after zone 2 while temperature (*T*) decreases uniformly as a function of distance (*D*) and sample length. The leftmost portion of zone 1 represents fully annealed material with grain growth and the rightmost area of zone 4 characterizes the as-received, mill-annealed condition. Here, zone 2 defines the area where recrystallization plus aging occurs and zone 3 depicts the area for recovery + aging reactions.

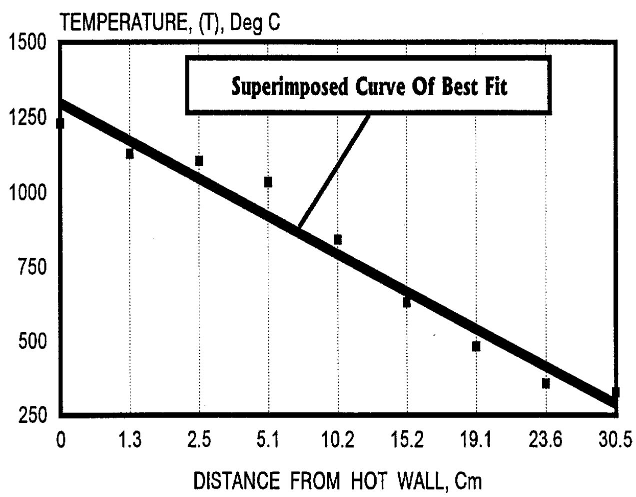


Fig. 1 Typical gradient-distance data for alloy C-350

Figure 4 defines the initiation stages for AR-aging: the effects of grain growth plus germination followed by recrystallization plus aging. Germination is defined as the "beginning to develop" stage.

Figure 5 is similar to Fig. 4, except that the rightmost end of Fig. 3 is better illustrated for the hardening of as-received, mill-annealed samples. Here, zone 3 identifies the region where recovery occurs simultaneously with aging, and zone 4 represents that part of the process where aging of the as-received and mill-annealed sample offers no significant increase in either hardness or strength.

If Fig. 3 to 5 are studied in detail, it is evident that an AR-aging heat treatment cycle requires an integration of different curves for grain growth, recrystallization, and recovery to be properly understood for this alloy system. These new findings provide the end user with a significant new processing challenge to incorrectly age harden an as-received, mill-annealed condition for less than optimum results or to correctly full-anneal a sample prior to optimized aging for enhanced results. This new heat treatment process is further described as a combination of these sequential events: annealing + recrystallization + aging.

Current metallurgical process practice, as controlled by military specification MIL-S-46850C, does not differentiate between the two important variables of mill-annealed versus full-annealed conditions. Further, this important specification does require aging of vacuum-melted material for 4 h at 482 °C (900 °F). This is to say that there is no apparent concern for different levels of stored energy from the producer mill for various thermal-reduction schedules that take place from ingot to billet to bar. In addition, this specification is unconcerned about another important process attribute for these particular alloys: to solutionize at a limit temperature of less than 1093 °C (2000 °F).

AR-aging requires that all as-received material from the producer mill be completely annealed to properly solutionize all of the hardening elements before aging is ever initiated. The effects of any prior aging and residual stresses are annihilated with an appropriate application of a solution annealing temperature. This treatment cycle leads to a better end result in full

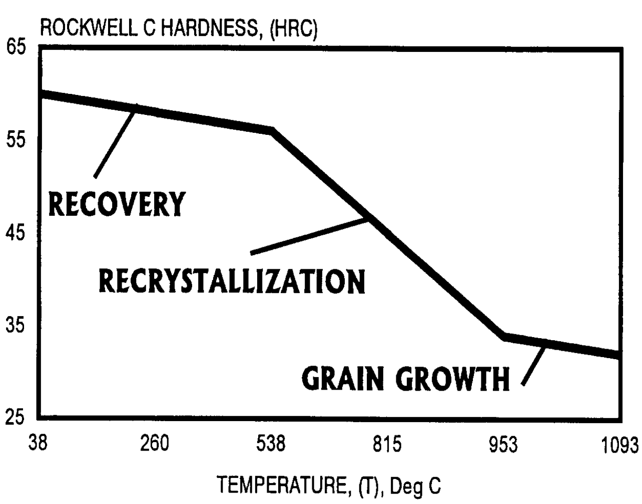


Fig. 2 Isochronal softening defined

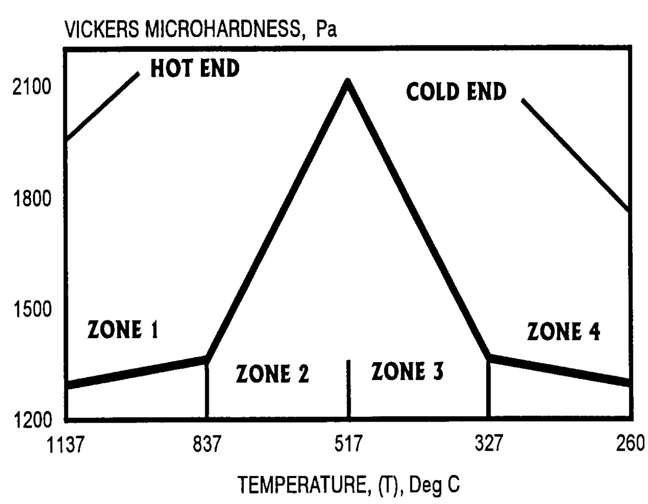


Fig. 3 Gradient profile for alloy C-350

agreement with those claims that were first reported by Adair et al. (Ref 6, 7). Table 2 cites recommended AR-aging treatment cycles to achieve a minimum grain size and an enhanced microstructure (without impurity segregations) for alloy C-350. When process variables of Table 2 are dutifully executed, interesting new results are achieved which have not been previously reported.

Figure (6) is an optical micrograph at an original magnification (M_0) of 400× to characterize the solution-annealed condition where a Vickers (HV) microhardness of 1343 Pa (329 kg/mm^2) was observed. Here, large grain diameters are evident without any residual, undesired effects caused by the mill-annealed and as-received condition. An optimized result has been here achieved for the solution-treated condition because this process temperature of 1057 °C (1935 °F) has not caused any segregation of impurities.

A set of new grains is shown in Fig. 7(a) for the recrystallized condition at an M_0 of 400×. When a solution-annealed sample is water quenched (WQ) from a temperature of 789 °C (1452 °F), the basic requirement for an optimum aging response is then executed; in other words, enough energy then exists to properly activate the subsequent sequence of age hardening.

This recrystallized condition is given as Fig. 7(b) at an M_0 of 1500×. Here, electron metallography reveals a small grain diameter for this soft as-quenched product, an HV of 1429 Pa (350 kg/mm^2). Again, there is no evidence of any blocky austenite and untempered martensite as undesired phase residuals from the as-received, mill-annealed condition. The objective of recrystallization has been here achieved; in other words, a new set of grains has been created, and the driving force for germination effects has been rendered for the subsequent aging treatment.

Figure 8(a) shows an optimized microstructure that exists when AR-aging is applied at a temperature of 517 °C (962 °F) for one hour followed by air cooling (AC) to ambient. A Vickers hardness of 3229 Pa (791 kg/mm^2) was observed without any residual effects from the as-received, mill-annealed state. This peak condition will offer a range in HV values that vary from about 3037 to 3229 Pa (744 to 790 kg/mm^2), depending on the aging time and the hardener composition. This corresponds to a range of about 62 to 64 HRC.

Electron metallography is given as Fig. 8(b) at an M_0 of 1500×. It is evident that the dissolution of untransformed austenite in alloy C-350 is a function of both low-angle and high-angle grain boundaries. This electron micrograph confirms that an AR-aging process is driven by a polygonization mechanism that involves a substructural strengthening event from proper processing of allied strain-anneal factors using reactions of recovery, recrystallization, and grain growth.

This new work does not attempt to confirm or deny any previous metallurgical theories about the hardening mechanism of MAR-aging steels through a martensitic transformation that is either diffusion-controlled or diffusionless with transformed austenite or irreversible gamma prime. This investigation does

Table 1 Nominal compositions of the Fe-base alloys studied

	Composition, wt%													
Type	Ni	Co	Mo	Ti	Cu	Al	Si	Cr	Mn	P	W	V	B	Zr
C200	18.2	7.5	4.4	0.2	0.14	0.1	0.05	0.04	0.01	0.005	0.01	0.01	0.003	0.012
C250	18.6	8.0	4.9	0.5	0.17	0.1	0.04	0.29	0.02	0.005	0.01	0.01	0.004	0.014
C300	18.6	9.2	4.8	0.7	0.17	0.1	0.06	0.19	0.01	0.004	0.01	0.01	0.005	0.017
C350	18.7	12.2	4.7	1.4	0.11	0.1	0.05	0.05	0.01	0.005	0.01	0.01	0.0033	0.012
T250	18.6	0.1	3.0	1.4	0.05	0.1	0.05	0.04	0.01	0.007	0.01	0.01	0.0001	0.01
T300	18.3	0.4	4.1	1.9	0.07	0.1	0.04	0.03	0.01	0.005	0.01	0.01	0.0002	0.01

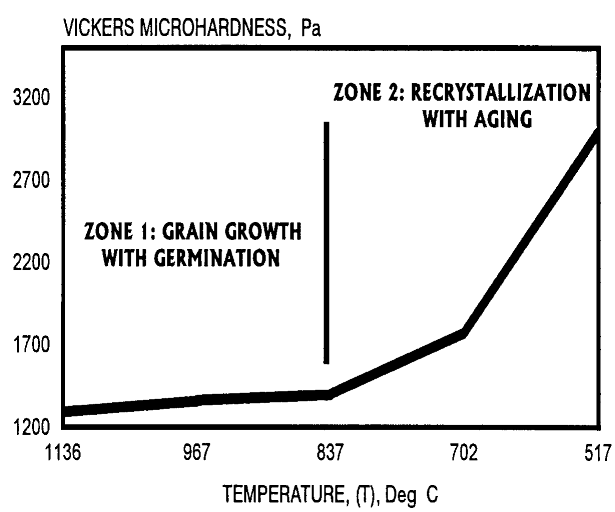


Fig. 4 High temperature end of gradient

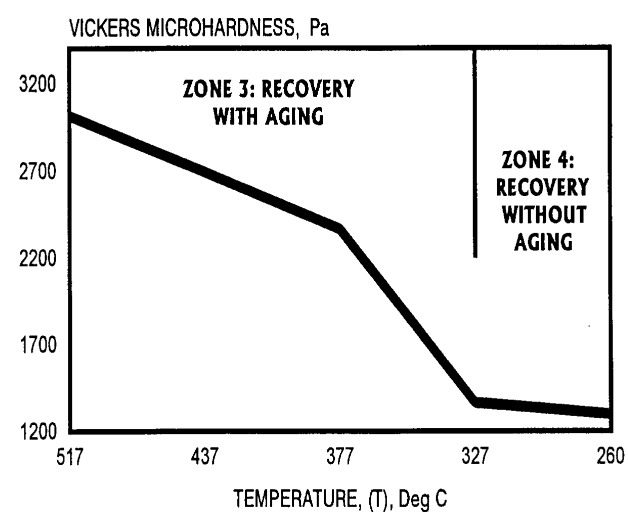


Fig. 5 Low temperature end of gradient

offer suitable evidence that this system of alloy steels can be effectively hardened using a new and different approach.

Figure 9(a) is an optical micrograph with an M_0 of 400× for a sample in the as-received, mill-annealed condition. Here, the presence of blocky austenite and untempered martensite is visible for a Vickers hardness of 1416 Pa (347 kg/mm^2).

Figure 9(b) is an electron micrograph at an M_0 of 1500× to document the fact that mill-annealed samples might undergo some recovery using heat treatment cycles within zone 4 without any concomitant effects from aging; in other words, no significant change in hardness and strength exists for samples that are so processed.

If Fig. 6 is compared to Fig. 9(a) (zone 1 versus zone 4), it is obvious that mill anneals and solution anneals offer decidedly different structures for similar hardness levels, 1343 versus 1416 HV (about HRC 34).

If differences between Fig. 7(a) and 9(a) are evaluated (zone 2 versus zone 4), it is apparent that recrystallization is the preference over mill-annealing for a similar hardness level of 1429 versus 1416 HV (about HRC 35). A set of new, small grains is a distinct advantage over the residual stresses and stored energy problems that dominate the mill-annealed state.

If zone 2 and zone 4 are again compared by a review of Fig. 8(a) and 9(a), it is evident that proper solution annealing plus recrystallization plus aging by the process of AR-aging can yield enhanced results. AR-aging can yield an optimized result of Vickers 3229 Pa (about HRC 64) while mill annealing plus recovery with aging can only warrant a minimum acceptable hardness of HRC 56 per MIL-S-46850C, which is equivalent to about Vickers 2551 Pa.

If this optimization effect is considered in terms of flow strength that is about equal to the conventional yield strength (Ref 8), samples can exhibit an enhancement of greater than 70,000 psi as compared to a conventional MAR-aged result (56 HRC compared against 64 HRC).

As first reported by Linnert (Ref 9), the alloy hardening of a given heat for a C-350 alloy composition can be estimated:

$$\Delta S_y \text{ (ksi)} = 8.8(\text{Co}) + 22.6(\text{Mo}) + 87.7(\text{Ti} + \text{Al})$$

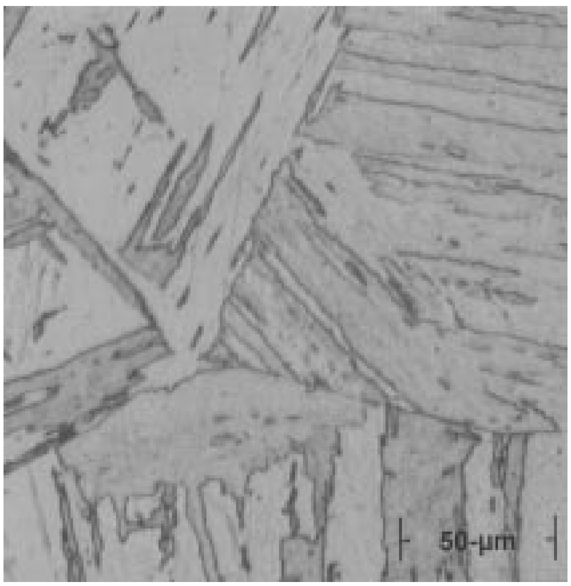
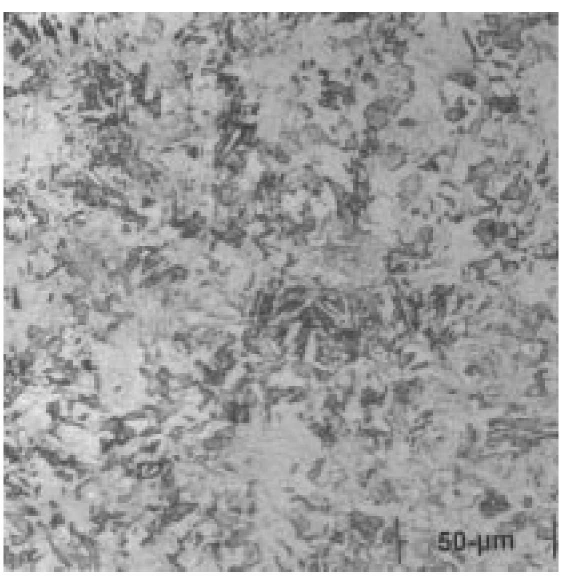


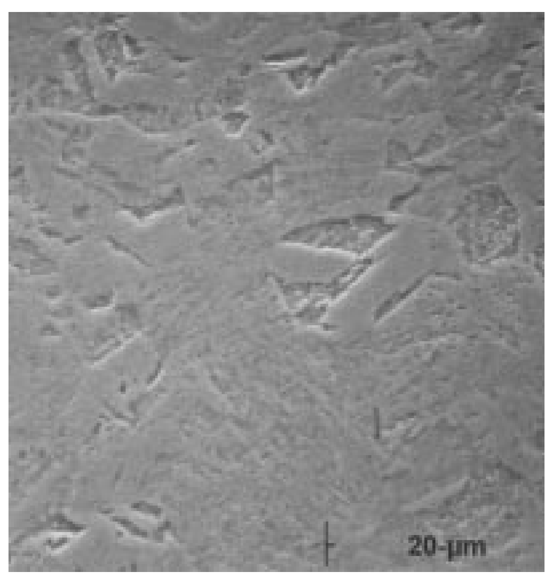
Fig. 6 Micrograph of fully annealed alloy C-350. 400×

Table 2 AR-aging of alloy C-350

Aging method	Temperature, °C (°F)	Time (min), h	Cooling method to ambient
Anneal	1057 (1935)	1	Air cool
Recrystallize	789 (1452)	1	Water quench
Age harden	517 (962)	1	Air cool



(a)



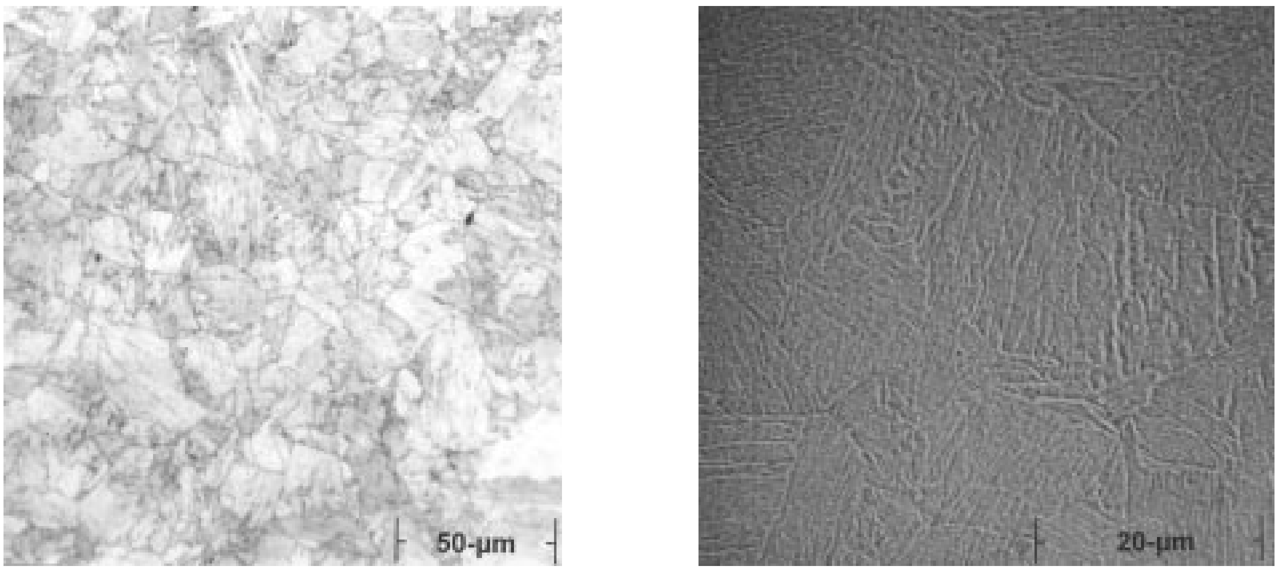
(b)

Fig. 7 Recrystallized alloy C-350 at (a) 400× and (b) 1500×

Table 3 Summary of important results

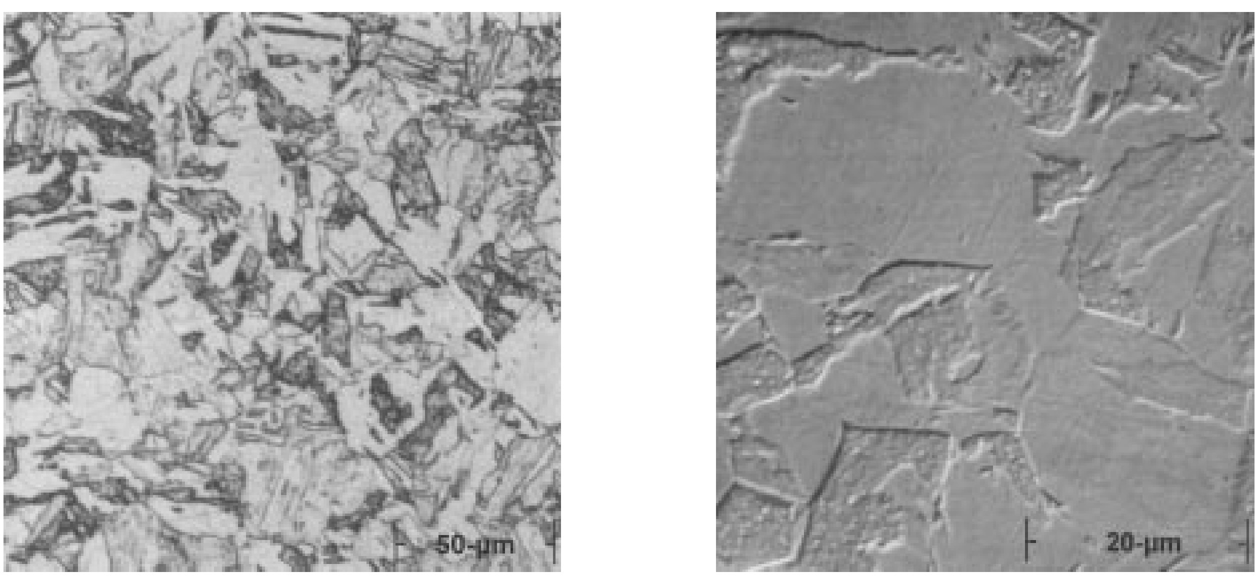
Metallurgical state	Isochronal process temperature, °C (°F)	Average Vickers microhardness, Pa (kg/mm^2)	Average Rockwell C hardness	Flow strength (S_F), MPa (psi)
Solution annealed	1057 (1935)	1343 (329)	33.3	1008 (146,000)
Recrystallized	789 (1452)	1429 (350)	35.8	1073 (155,550)
Recrystallized and aged	517 (962)	3229 (791)	64.2	2424 (351,575)
Mill-annealed	260 (500)	1416 (347)	35.6	1063 (154,000)

Minimal acceptable hardness for MAR-aged C-350 alloy is 56 HRC per MIL-S-46850C, which is equivalent to about 2551 Pa (625 kg/mm^2). Rockwell C hardness data were determined from conversion charts for MAR-aging steel. Flow strength (S_F) was calculated from Tabor's law (Ref 8).



(a) (b)

Fig. 8 AR-aged alloy C-350 at (a) 400× and (b) 1500×



(a) (b)

Fig. 9 Mill-annealed alloy C-350 at (a) 400× and (b) 1500×

On this basis and without regard to any size effects and/or microsegregations this heat (see Table 1) would exhibit a maximum yield strength (S_y) of 2379 MPa (345 ksi). Table 3 shows that AR-aging can produce a flow strength of 2424 MPa (351.5 ksi) so these two strength values are similar. Assuming that the minimum acceptable flow strength for a MAR-aged product is 1915 MPa (277.8 ksi) per MIL-S-46850C (based on 56 HRC), then the percent difference between two important design levels for C-350 (2424 MPa versus 1915 MPa) is approximately a 24% difference, which is statistically significant. This is to say that AR-aging can offer much improvement over what is often called minimum requirements (56 versus 64 HRC).

If Fig. 7(b) and 9(b) are evaluated, the primary difference to be noted is that one process treatment (recrystallized) leads to a fine grain size practice, while the traditional treatment of mill-annealing with recovery causes a coarser grain size. And, for this alloy of C-350, fine grain size approaches are always required if life extension is of concern for noncreep applications.

If Fig. 8(b) and 9(b) are studied, the ultimate difference for AR-aging is here depicted through the controlling force of polygonization in lieu of transformation effects caused by conventional MAR-aging.

5. Conclusions

This investigation has been sufficient to offer these summary remarks:

- MAR-aging steels do respond in a significant manner to conventional recovery, recrystallization, and grain growth processing by a new process called AR-aging.
- The reactions of solution annealing and age hardening are reversible reactions if the hardening elements are redissolved at a limit temperature of less than 1093 °C (2000 °F), and if recrystallization-aging follows a satisfactory solutionization treatment. This confirms earlier work by Adair et al. (Ref 7) who claimed that an elevated thermal treatment is mandated if MAR-aging steels are to achieve an optimized austenitic grain size for improved mechanical/metallurgical properties. Adair's high-temperature treatment for grain refinement has not been commonly used during the intervening years.
- Of the two processing treatment methods (MAR-aging versus AR-aging), AR-aging is preferred because it virtually eliminates the problem of undesired residual phases in this strategically vital family of Fe-18Ni alloy steels.
- AR-aging procedures have proven to be of extreme importance for many different applications, but two recent instances quickly come to mind: a component for a nuclear reactor application in the Northwest and precision-made Hopkinson bars to be used by prestigious universities of the East and in the United Kingdom. Hopkinson bars are used for reference purposes where ever so slight changes in size are of utmost importance. In both cases, the original mill-annealed condition was neither acceptable nor usable, so AR-aging techniques had to be applied in lieu of conventional MAR-aging procedure, and this process change to AR-aging did yield successful results.

References

1. C.G. Bieber, Progress with 25% Nickel Steels for High Strength Applications, *Met. Prog.*, Nov 1960
2. S. Floreen and R.F. Decker, Heat Treatment of 18% Ni MAR-Aging Steel, *Trans. ASM,* Vol 55, 1962
3. R.F. Decker, Notes on the Development of MAR-Aging Steels, *Source Book on MAR-Aging Steels*, American Society for Metals, 1979, p xi-xv
4. R.F. Decker, J.T. Eash, and A.J. Goldman, 18% Nickel MAR-Aging Steel, *Trans. ASM,* Vol 55, 1962, p 58
5. H. Margolin, *Recrystallization, Grain Growth and Textures,* American Society for Metals, 1966
6. G.E. Pellissier, *Eng. Fract. Mech.,* Vol 1, 1968, p 65
7. G. Saul, J.A. Roberson, and A.M. Adair, The Effects of Thermal Treatment on the Austenitic Grain Size and Mechanical Properties of 18% Ni MAR-Aging Steels, *Metall. Trans.,* Vol 1, Feb 1970
8. D. Tabor, *The Hardness of Metals,* Clarendon Press, 1951, p 104
9. G.E. Linnert, *Welding Metallurgy,* Vol 2, 3rd ed., AWS, 1967, p 528