THE PHYSICAL METALLURGY OF A SILICON-CONTAINING LOW EXPANSION SUPERALLOY

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INCOLOY* alloy 909 is an iron-nickel-cobalt-niobium-titanium controlled low thermal expansion superalloy with an intentional silicon addition. Recommended for use at temperatures to 650°C, the alloy is now specified for major new gas turbine engines. The pseudo-equilibrium time - temperature - transformation behavior of alloy 909 is presented as well as a brief description of the major phases: γ', ε'', ε, and Laves. Gamma prime (γ'), the major strengthening phase in the alloy, is shown to precipitate in the 538°C to 760°C range. The ε'' and ε phases, which are similar to γ'' and δ phase found in INCONEL* alloy 718, precipitate at intermediate temperatures (about 700°C to 950°C). Laves phase, which is used to control grain size, precipitates at the higher temperatures (800°C-1040°C) used during hot working and annealing. The alloy's Si addition significantly affects the formation of these phases during processing, thereby affecting mechanical properties. The interrelationship between the physical metallurgy and properties of the alloy are explored in a two level factorial study of processing (two levels of Laves precipitation) and age hardening heat treatments (the alloy's standard heat treat cycle and a short time cycle). Notched bar rupture tests and creep crack growth tests at 538°C demonstrate that resistance to stress accelerated grain boundary oxygen embrittlement (SAGBO) improves with precipitation of intergranular ε phase. Excess Laves precipitation lowers residual Nb content, consequently reducing ε abundance and reducing crack growth resistance. Combined with proper thermomechanical processing, the short aging cycle precipitates an ε grain boundary structure which significantly improves SAGBO resistance.

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Introduction

INCOLOY alloy 909 is the latest development in a series of low expansion superalloys designed for use up to 650°C where age-hardened strength and close operating tolerances are required. Introduced at the Fifth International Symposium on Superalloys (1), this alloy is now specified in major new gas turbine engines and is being evaluated for many other applications.

Like other controlled expansion superalloys, alloy 909 derives its low expansion characteristics from electron spin interactions within its Fe-Ni-Co matrix. The high strength of these alloys is achieved by the precipitation of A_3B type phases, where B may be Al, Ti, or Nb, though INCOLOY alloys 907 and 909 rely only on Ti and Nb for strengthening. The necessity of a Cr-free matrix to achieve low controlled expansivity results in reduced oxidation resistance and a phenomenon known as stress accelerated grain boundary oxygen embrittlement (SAGBO), which occurs when material is stressed at intermediate temperatures in air.

In the earliest of these alloys, INCOLOY alloy 903, the threshold stress for oxygen embrittlement was extremely low except in the longitudinal direction of heavily warm-worked material (2). The restricted Al and increased Nb content of INCOLOY alloy 907, combined with an overaging heat treatment raised this threshold stress considerably (1,2). In alloy 909, the addition of 0.4% Si combined excellent SAGBO behavior with good tensile properties and eliminated the extreme processing and/or heat treatment measures of the earlier alloys. Substantial and complex differences in microstructure and physical metallurgy accompanied this small Si addition.

In this two part paper, Part I describes the alloy's time-temperature-transformation (TTT) behavior and its major precipitated phases. Part II applies this understanding to a two-level factorial study of thermomechanical processing and age hardening heat treatments.

I. Isothermal Time-Temperature-Transformation Study

Experimental Procedures

The TTT study was conducted on a commercially produced hot rolled flat containing (by weight %) 42.1% Fe, 38.3% Ni, 12.9% Co, 4.70% Nb, 1.58% Ti, 0.35% Si, 0.04% Al, 0.01% C, 0.01% Cr. Specimens were solution annealed at 1038°C for one hour, air cooled, then isothermally heat treated in 56°C increments from 538°C through 1038°C for 0.1, 1, 10, 100, and 1000 hours.

Bulk heats and extracted residues were chemically analyzed by X-ray spectrography and inductive-coupled plasma (ICP) analysis. Point probe analyses were performed on a microprobe and a SEM with EDX. Thin foils and extraction replicas were also examined by TEM at 100kV. X-ray diffraction (XRD) analyses of extracted phases were conducted on an automated diffractometer system using copper radiation. Previously unknown diffraction patterns were indexed by methods described by Cullity (3).

Conventional specimen preparation techniques are not suitable because the alloy is ferromagnetic, has poor oxidation resistance, and contains phases with modified structures. Because of space limitations, the new preparation and analysis techniques are not detailed here.

Review of Phase Analyses

Overview. Figure 1, the TTT diagram for solution annealed alloy 909, shows the precipitation C-curves for the intragranular γ', ε'', ε phases and the grain boundary (noted as G.B.) phases of the Laves and ε types. G-phase silicides ($A_{16}B_6Si_7$) form in the Laves region after prolonged exposures (>100 hours) (8). Minor phases present, but not shown, include (Nb,Ti)(C,N), TiN and $Ti_2C_4S_2$.

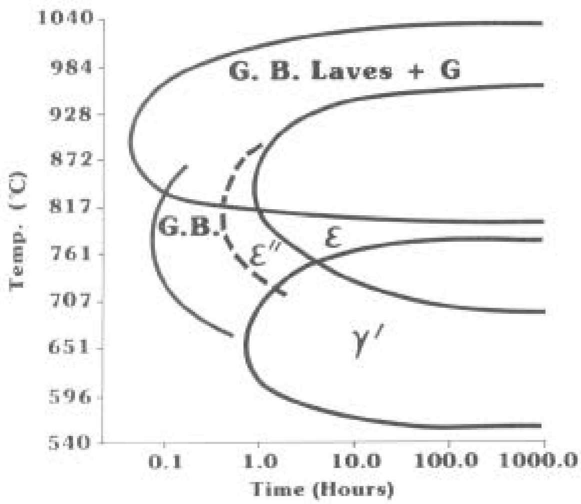

Figure 1: TTT diagram for INCOLOY alloy 909 (0.35% Si). Isothermal aging after 1038°C/1 hour anneal.

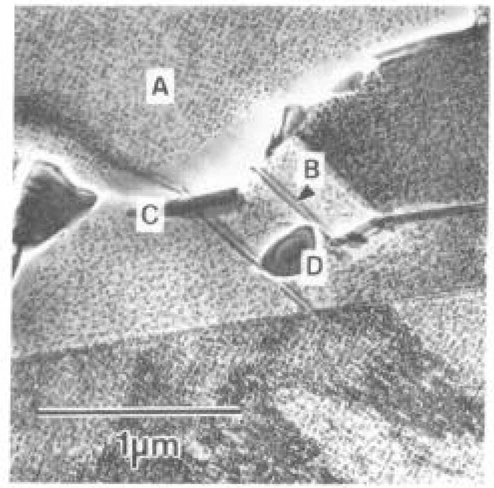

Figure 2: Thin foil TEM image after standard treatment (see Part II). A= $\gamma+\gamma'$, B= ε'', C= ε, D= Laves.

Table I (located at the end of Part I) gives X-ray diffraction data for extracted major precipitates which have the following approximate compositions (in atomic %):

Phase	Fe	Ni	Co	Nb	Ti	Other
γ'	8	51	12	13	16	---
ε''	12	46	12	14	16	---
ε	11	50	16	18	4	2 Si
Laves	18	38	14	18	3	5 Si,2 C

Gamma prime (γ'). This major strengthening constituent precipitates between 538°C and 760°C, and is predominantly $Ni_3(Ti,Nb)$ with an $L1_2$ structure. The fact that γ'' (a transition phase in other Nb strengthened alloys such as alloy 718) was not found is likely due to the effect of Co and Fe on stacking fault energy. The transmission electron photomicrograph in Figure 2 shows intragranular γ' along with the ε'', ε and Laves phases precipitated during the alloy's standard heat treatment (see Part II).

Epsilon double prime (ε''). At higher temperatures (700°C-950°C) a $(Ni,Fe,Co)_3(Nb,Ti)$ transitional phase of very fine inter- and intragranular platelets precipitates. Together, the diffraction patterns for ε'' and Laves phases (Table I) can be mistaken as delta (δ) phase. The ε'' phase also occurs in commercially overaged alloy 907, but the Si content of alloy 909 accelerates its precipitation rate and broadens its temperature range.

The acicular morphology of ε'', shown in Figure 3, is similar to δ or γ''. (This accentuated precipitation is shown in warm-worked and aged material.) While its exact structure is unknown, it is expected to be distorted hexagonal or FCC compatible with the transition of γ' (abcabc) to the equilibrium ε (abab) phase.

Epsilon (ε). This phase initially precipitates at grain boundaries, around nitrides and carbonitrides, and intragranularly from ε". The angular, blocky or acicular intergranular and Widsmanstatten intragranular precipitates appear optically similar to δ phase in alloy 718. As with ε", Si enhances the precipitation of this phase. Whether ε precipitation is always preceded by ε" is uncertain. Figure 4 shows the platelet morphology of ε, which has a DO_{19} hexagonal superlattice (Ni_3Sn-type) structure. The Co content of ε is greater and the Ti content lower than shown for ε". Calculations from composition data of ε phase fall neatly into the DO_{19} structure field as defined by Watson and Bennett(4). Eta (η), a DO_{24} structure of Ni_3Ti with a similar morphology to ε, was not found.

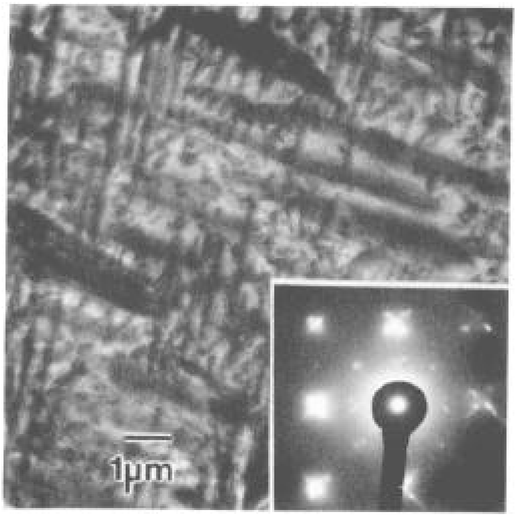

Figure 3: Thin foil TEM images of ε". Note similarity of pattern to <001> FCC γ', and heavy streaking causing displacement of superlattice spots.

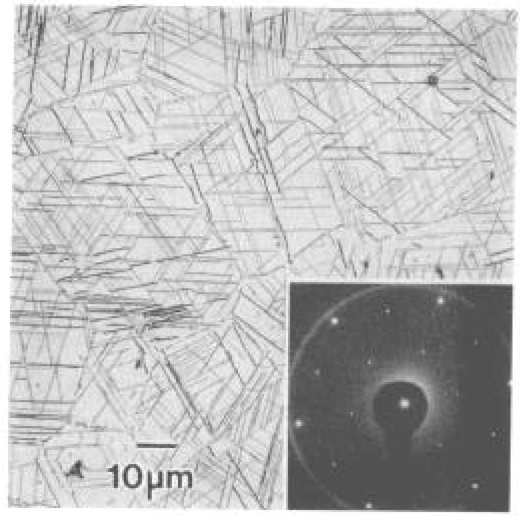

Figure 4: Optical micrograph of ε in an overaged sample. TEM diffraction pattern of extracted platelet is DO_{19} <001>. Ring is 0.204nm Cr.

Laves. Two and four-layered hexagonal Laves phases ($MgZn_2$ and $MgNi_2$ type crystal structures) form in grain boundaries at the high temperatures (800°C-1040°C) routinely encountered in hot working and annealing. The Si addition in alloy 909 induces the Laves precipitation by increasing the N_v of the matrix and stabilizing TCP phases (5). This effect is profound since Laves has not been observed in the lower Si alloy 907.

These Ni-rich phases have similar lattice parameters to Laves found in the Co-Nb binary system (6,7). Point-probe analyses of extracted Laves particles did not indicate any notable compositional differences. However, sample to sample variations in the d-spacings of the four-layered phase suggested slight differences could exist. The lattice parameters of the two-layered phase were equivalent from sample to sample. The two phases probably exist in varying ratios within a single, faulted particle.

Kinetic and Compositional Factors. In commercial practice, two effects modify the TTT behavior in Figure 1. First, the precipitation kinetics of the major phases are sensitive to internal energy levels. C-curves of material containing residual work or annealed at lower temperatures will be significantly displaced to shorter times with broadened temperature ranges. Second, the major phases can be significantly affected by variations in minor element content, especially Si, as well as the major elements.

Phase-property-SAGBO interrelationships. A primary concern in controlled expansion superalloys is oxygen embrittlement (SAGBO). Alloy 909 achieves its degree of SAGBO resistance by the formation of a fine grained structure and the precipitation of ε and perhaps ε" phases.

Grain refinement is achieved by controlling Laves quantity and distribution during thermomechanical processing. Sufficient Laves must be precipitated to permit grain boundary pinning, but very coarse Laves particles decrease the grain refining effect. A heavy Laves prior grain boundary network may contribute to reduced ductility. Excessive quantities are also undesirable as lower residual Nb may reduce ε and ε" phase precipitation during subsequent heat treatment. The resulting change in the Nb/Ti ratio may affect intragranular overaging rate and properties by altering coherency strains of strengthening precipitates.

Although the mechanism(s) is not fully understood, the presence of ε, and probably ε", in existing grain boundaries is required to offset oxygen embrittlement (as grain boundary δ benefits the rupture properties of alloy 718). The abundance and temperature regimes of these phases are sensitive to both composition and strain energy in the material before aging. Residual strain energy in the presence of Si promotes the precipitation of the beneficial intergranular phases and allows an excellent combination of SAGBO resistance and tensile properties to be achieved in commercially attractive aging cycles.

TABLE I - INCOLOY alloy 909: X-ray Diffraction Data - Phase Extractions

MAJOR PHASES														MINOR PHASE	
Matrix		Matrix (Aged)		Gamma Prime		Epsilon Dbl.Prime		Epsilon		Laves Phases 2-layer		Laves Phases 4-layer		G	
a=.3608		a=.3605		a=.3627		a=.518? c=.420?		a=.5177 c=.4196		a=.4770 c=.7759		a=.4810 c=1.589		a=1.125	
d(nm)	I	d(nm)	I	d(nm)	I	d(nm)	I	d(nm)	I	d(nm)	I	d(nm)	I	d(nm)	I
												.405	2		
												.332	4	.325	16
								.306	3						
												.297*	21		
										.282	10				
				.2560	48									.256	6
												.241	27		
										.238	40				
												.229*	17		
						.223	17	.223	25						
										.219	52				
														.216	100
.2083	100	.2081	100	.2084	100	.209	100	.2096	+80						
										.206	6	.205	18		
										.203	100				
						.201*	70					.201	26		
										.1996	60	.1990	100		
						.196	90	.1972	100					.198	100
						.193*	35			.194	10	.1926	7	.190	9
.1804	20	.1802	20	.1806	50									.187	16
								.1527	15						
												.1401	8		
												.1356	4		
												.1327	5	.1326	12
.1276	21	.1275	21	.1278	48	.1292	23	.1293	15	.1298	10			.1303	6
										.1241	8	.1234	2		
								.1185	19					.1131	10
.1089	20	.1087	20	.1090	38	.1100	18	.1099	21					.1100	4
								.1080	12					.1088	5
.1042	7	.1041	7	.1043	6	.1051	3	.1047	3						
.0828	14	.0827	14	.0832	19										
				.0811	25										

+ High intensity due to texture. * Peaks do not match calculated pattern.

II. Process - Aging Treatment Factorial Study

Procedure - Factorial Study

Experimental Design. To demonstrate the fundamental uses of the phases described in Part I and to illustrate their resulting effects on mechanical properties, it was decided to examine the interactive effects of processing and heat treatment using a two by two factorial design. Two hot working processes providing two levels of Laves phases were combined with two aging cycles.

Hot Working. The starting material for both experimental processes was 90 mm ϕ hot rolled and rough turned bar from a commercial melt with an average chemistry of 41.4% Fe, 38.4% Ni, 13.0% Co, 4.84% Nb, 1.65% Ti, 0.41% Si, 0.04% Al, 0.005% C, 0.14% Cr. Bars were rolled to a final size of 14 mm thick by 64 mm using two Processes: A and B.

In Process A, the bar was heated for one hour (1 h) at 1038°C and rolled to 41 mm square, reheated for 0.5 h at 1024°C, flat rolled to 20 mm thickness ($\geq$15% reduction below 927°C), reheated for 0.5 h at 996°C and rolled to final size ($\geq$15% reduction below 927°C).

In Process B, the bar was heated for 1 h at 996°C and rolled to 41 mm square, reheated for 2 h at 968°C, flat rolled to 32 mm thickness, reheated for 2 h at 968°C, rolled to 22 mm thickness, reheated for 2 h at 968°C and rolled to final size ($\geq$25% reduction below 927°C). The additional reheat, lower heating temperatures and extended soaking times for this process were selected to produce excessive Laves and reduce residual Nb content.

Heat Treatment. Test blanks were given a recrystallizing, fine grained anneal of 982°C for 1 h and air cooled (AC). Unlike the 1038°C treatment used for the TTT study, this anneal does not solution the Laves phase(s) but instead precipitates Laves in a prior grain boundary network. The retained energy from this lower annealing temperature accelerates the subsequent precipitation of intermediate temperature phases when aging with one of the following treatments (noted as STAND and SHORT):

STAND - 718°C/ 8 h furnace cooled (55°C/ h) to 621°C/ 8 h, AC.
The alloy's standard age gives an excellent combination of tensile and 649°C stress rupture strength and is widely used with alloy 718 (1).

SHORT - 746°C/ 4 h furnace cooled (55°C/ h) to 621°C/ 4 h, AC.
Earlier unreported work showed this treatment as an economical, yet effective age. Some sacrifice of tensile properties resulted, but rupture properties were quite good. Also, the TTT diagram suggested that a higher initial aging temperature (albeit shorter time) could promote more precipitation of ε and ε'', thought beneficial to SAGBO resistance.

Testing Procedures. The following mechanical property tests were conducted: room temperature tensile (20°C), 649°C high temperature tensile, 649°C, 510 MPa combination smooth and K_t 3.6 notched stress rupture bar and 538°C, 827 MPa K_t 2 notched stress rupture bar. (All smooth section gauge lengths were four times the gauge diameter. Smooth gauges and notches for high temperature tests were finished using a low stress grinding technique.)

Duplicate crack growth tests (9.5 mm thick compact tension specimens) were conducted in air at 538°C using both static and fatigue loading. Notches were oriented parallel to the rolling direction. Fatigue pre-cracking and crack growth testing were conducted in accordance with ASTM A647-85. Crack lengths were measured optically.

The K-increasing fatigue crack growth tests were conducted at Metcut Associates under constant peak load and amplitude using a simple linear ramp waveform at a frequency of 0.033 Hz and R ratio of 0.1.

Data Review

Mechanical Properties. Examination of processing and aging effects in factorial Table II shows:

1. Yield and tensile strengths were only slightly affected by processing. For Process B (excess Laves) strengths were lowered $\leq$ 1% at 20°C and $\leq$ 5% at 649°C. As expected, strengths were reduced by the short-time age. In this case, the decrease in yield and tensile strengths ranged from 7-10% and 4-8% respectively. Tensile ductilities were virtually unaffected by either variable.

2. All of the 649°C, 510 MPa combination smooth and notched bar stress rupture tests were notch ductile (i.e., broke in the smooth gauge) and showed excellent life and ductility. Given the normal variation in stress rupture testing there seemed to be little, if any, effect of processing on rupture life, and aging with the short cycle showed a mild negative effect at worst.

3. The 538°C notch results confirmed earlier findings (1) which demonstrated this test to be more sensitive to SAGBO than the 649°C test. Notch lives varied by an order of magnitude (i.e., from about 40 h to over 400 h). There was little effect of processing with the standard aging cycle but there was a strong, positive interaction with Process A (normal Laves) and the short-time age.

Crack Growth Results. The static crack growth (SCG) rates in Figure 5 were lowest in specimens given the combination of Process A and the short-time age. Rates were highest in Process B specimens given the standard age. The strong correlation between 538°C notch rupture and SCG behavior, coupled with their similar intergranular crack appearance (not shown), indicates the key to improving notch properties is decreasing SCG rate.

As discussed below, it is necessary to control processing to promote subsequent ε precipitation. Process control should include both grain size and Laves precipitation controls.

In contrast to SCG, fatigue crack growth (FCG) rates were relatively unaffected by process or heat treatment indicating that crack growth is dependent on the time under applied stress. One expects that fatigue profiles containing hold times at peak stress would show similar microstructural effects on FCG rate as observed on SCG behavior.

Microstructure Review. As expected, the two processes produced different amounts of Laves and age hardening precipitates. Extractions of the annealed Process A material contained 1.5 wgt% Laves compared to 3.5 wgt% for Process B, and EDX analysis of grain interiors showed a corresponding reduction of residual Nb with the latter process. Regardless of the aging cycle, extractions of fully heat treated materials showed Process A contained 6.1 wgt% A_3B age hardening phases vs. 4.1 wgt% for Process B.

Although it was intended that Processes A and B yield equal grain sizes, Process B material had an ASTM#9 grain size compared to Process A's ASTM#7.5 structure. Grain refinement has a strong positive effect on the SAGBO/ notch rupture strength of these alloys (2). Thus, on the basis of grain size alone, Process B should have had an edge over Process A.

TABLE II - INCOLOY alloy 909: Effect of Process and Aging Treatment on Mechanical Properties

		Age: STAND		Age: SHORT	
		Process:		Process:	
		A	B	A	B
20 C Tensile	YS (MPa):	1102	1089	998	996
	TS (MPa):	1366	1353	1309	1298
	El (%):	18	17	18	18
	RA (%):	40	42	42	44
649 C Tensile	YS (MPa):	931	888	832	826
	TS (MPa):	1093	1067	1009	984
	El (%):	16	22	21	23
	RA (%):	52	60	61	59
Comb. Rupture 649 C 510 MPa	Life (hr):	141	118	101	110
	El (%):	30	33	18	30
	RA (%):	48	57	30	41
Notch Rupture 538 C 827 MPa	Life (hr):	62*	80	417	100

* - Average of 41, 53, and 91 hrs.

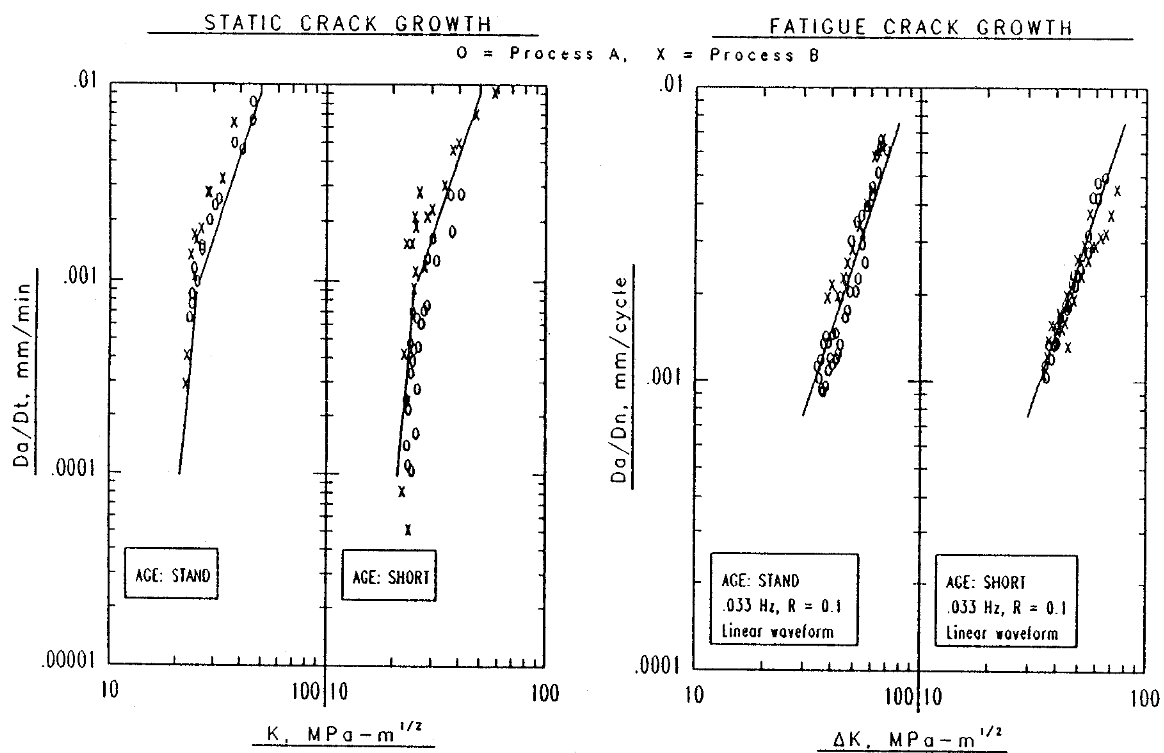

Figure 5. Effect of process and aging treatment on 538°C static and fatigue crack growth of INCOLOY alloy 909. The overall mean curves for both SCG and FCG are shown for comparison purposes.

Despite its coarser grain size, Process A material given the SHORT age showed substantial improvements in 538°C notch rupture and static crack growth behavior. This indicates that the location, quantity or morphology of age hardening precipitates are important factors in obtaining superior SAGBO resistance. Figures 6 and 7 show differences in grain boundary ε precipitation of Process A material given the two aging treatments. These optical and TEM photomicrographs demonstrate that the standard treatment has a less continuous, more globular precipitate compared to the more complete zipper-like acicular morphology found in the SHORT age material . The higher residual Nb content in Process A apparently shifts the ε C-curve to shorter times thus allowing more ε precipitation to occur. The 28°C higher intermediate aging temperature of the SHORT cycle more than compensates for its shorter time. Together, Process A and the SHORT cycle were very effective in accelerating ε precipitation, changing its distribution and morphology, and improving SAGBO performance. Unreported work showed a further increase in intermediate aging temperature would result in minimal improvement in SAGBO resistance compared to the loss in strength.

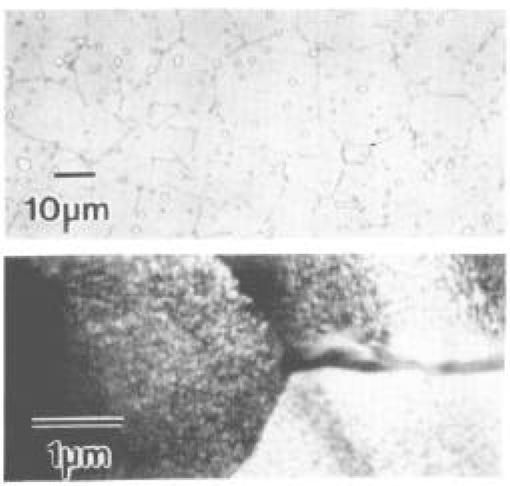

Figure 6: Optical (a) and TEM (b) micrographs of Process A material after STAND age.

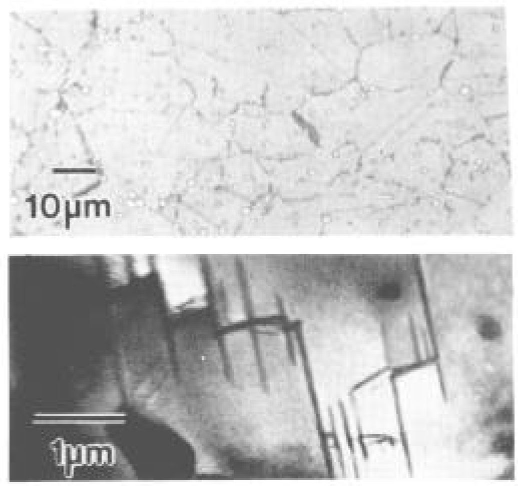

Figure 7: Optical (a) and TEM (b) micrographs of Process A material after SHORT age.

Summary

This investigation reveals the TTT behavior of INCOLOY alloy 909, an age hardenable Fe-Ni-Co material with a low coefficient of thermal expansion. These findings are applied in the design of a factorial experiment to examine the effects of processing and heat treatment on the mechanical properties and crack growth behavior of the alloy. Major findings include:

1. Gamma prime (γ′), the alloy's primary strengthening phase, precipitates at temperatures below about 760°C and consists of $Ni_3(Ti,Nb)$.

2. Epsilon double prime (ε″) and epsilon (ε) are transitional and equilibrium A_3B phases which precipitate during aging at intermediate temperatures. While ε″ and ε resemble the Ni_3Nb delta (δ) phase found in alloy 718, they are crystallographically distinct. Together, the grain boundary forms of these phases improve the alloy's resistance to stress accelerated grain boundary oxygen embrittlement (SAGBO).

3. Laves phases can precipitate in a prior grain boundary network at the higher temperatures used in commercial hot working and annealing. With proper process controls, Laves is essential to the grain refinement contributing to SAGBO resistance.

4. There are strong effects of matrix composition on TTT behavior:
   a.) Si raises the alloy's N_v which promotes the formation of Laves phases, and broadens the TTT curves for ε" and ε.
   b.) Fe and Co affect the ordering of the A_3B phases and favor formation of ε-type structures (instead of γ" and δ).
   c.) Excessive Laves precipitation reduces Nb in the matrix, affecting subsequent formation of beneficial ε-type phases.

5. The factorial study of processing and aging treatment showed tensile properties were little affected by processing. The short age cycle decreased yield and tensile strengths up to 10%, but combined with Process A (normal Laves), this age gave excellent 538°C SAGBO resistance (as evidenced by notch rupture and static crack growth results).

6. The dramatic improvement in SAGBO strength was accomplished by selecting a process/ heat treatment combination which created a beneficial microstructure containing both the grain-refining Laves phase and the grain boundary ε phase.

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