Cross-Roll Flow Forming of ODS Alloy Heat Exchanger Tubes For Hoop Creep Enhancement

Quarterly Technical Progress Report
Jan 1st 2005 – Mar 31st 2005

Principal Investigator:

Bimal K. Kad
Department of Structural Engineering
University of California-San Diego, La Jolla, CA 92093-0411
Tel: (858) 534 7059; Fax: (858) 534-6373; e-mail: bkad@ucsd.edu

Issue Date June 27th 2005

National Energy Technology Laboratory
Contract Award: DE-FC26-03NT41985

For

Development of Technologies and Capabilities for Developing Coal, Oil and Gas Energy Resources

Cross-Roll Flow Forming of ODS Alloy Heat Exchanger Tubes For Hoop Creep Enhancement

Abstract

Mechanically alloyed oxide dispersion strengthened (ODS) Fe-Cr-Al alloy thin walled tubes and sheets, produced via powder processing and consolidation methodologies, are promising materials for eventual use at temperatures up to 1200^{o}C in the power generation industry, far above the temperature capabilities of conventional alloys. Target end-uses range from gas turbine combustor liners to high aspect ratio (L/D) heat exchanger tubes. Grain boundary creep processes at service temperatures, particularly those acting in the hoop direction, are the dominant failure mechanisms for such components. The processed microstructure of ODS alloys consists of high aspect ratio grains aligned parallel to the tube axis, a result of dominant axial metal flow which aligns the dispersoid particles and other impurities in the longitudinal direction. The dispersion distribution is unaltered on a micro scale by recrystallization thermal treatments, but the high aspect ratio grain shape typically obtained limits transverse grain spacing and consequently the hoop creep response. Improving hoop creep in ODS-alloy components will require understanding and manipulating the factors that control the recrystallization behavior, and represents a critical materials design and development challenge that must be overcome in order to fully exploit the potential of ODS alloys.

The objectives of this program are to 1) increase creep-strength at temperature in ODS-alloy tube and liner components by 100% *via*, 2) preferential cross-roll flow forming and grain/particle fibering in the critical hoop direction. *Recent studies in cross-rolled ODS-alloy sheets (produced from flattened tubes) indicate that transverse creep is significantly enhanced via controlled transverse grain fibering, and similar improvements are expected for cross-rolled tubes*. The research program outlined here is iterative in nature and is intended to systematically i) prescribe extrusion consolidation methodologies via detailed test matrices, ii) examine and identify post-extrusion forming methodologies to create hoop strengthened tubes, which will be iii) evaluated at 'in-service' loads at service temperatures and environments. This research program is being conducted in collaboration with the DOE's Oak Ridge National Laboratory and the vested industrial partner Special Metals Corporation. In this sixth quarter of performance, program activities are continued for Tasks 2, 3 and 4 and are reported herein. The creep performance enhancement in cross-rolled MA956 materials samples versus the base creep property is elucidated. *At least 1-2 orders of magnitude of improvement in creep rates/day are demonstrated for the cross-rolled samples versus the base reference tests*. Furthermore, it appears that 20% cross-rolling stain is sufficient to create optimum strengthening, as larger strains achieved in flow formed materials yield no additional hoop creep enhancement.

Cross-Roll Flow Forming of ODS Alloy Heat Exchanger Tubes For Hoop Creep Enhancement

Table of Contents

§ 1. Executive Summary

Oxide dispersion strengthened (ODS) ferritic alloys based on FeCrAl and intermetallic Fe_3Al alloys are promising materials for high-temperature, high-pressure tubing, liner and shell applications on account of their creep strength at very high temperatures and excellent corrosion resistance in oxidizing, oxidizing/sulphidizing and oxidizing/chlorinating environments compared to available high-temperature alloys. Requirements for such a combination of properties are found in advanced systems being developed for utilization of fossil fuels, such as the **DOE**'s **Vision 21** and **FutureGen** programs and in improved gas turbines being developed for power generation.

The creep strength of conventional high-temperature alloys decreases rapidly with increasing temperature, as shown in Fig. 1, since the thermodynamic stability of the various available strengthening phases also decreases with increasing temperature[1]. Also shown in Fig. 1 is the significant increase in temperature capability afforded when a dispersion of inert oxide particles is used as the strengthening phase. A major feature of oxide dispersion-strengthened alloys is that the most successful route for their preparation appears to involve powder metallurgical processing[2-6]. Further, the critical need to maintain the fine size, volume fraction, and uniform distribution of the oxide particles in the alloy matrix, as well as the need to develop specific grain shapes, results in some significant differences in alloy fabricability and in the application of joining procedures, compared to conventional cast and wrought alloys. Hence, while ODS alloys offer a significant increase in temperature capability, they have a limited formability envelope, their mechanical properties are very anisotropic, and they cannot be joined by conventional fusion welding processes. Thus, the exploitation of the full capabilities of ODS alloys is limited until these critical hurdles are addressed and overcome.

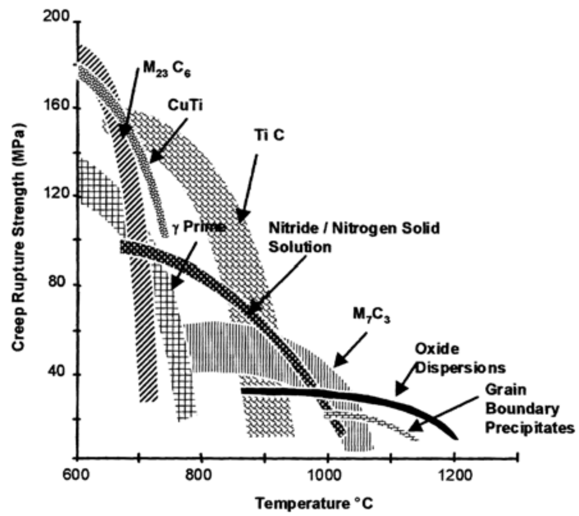

Figure 1. The creep performance envelope as a function of strengthening phase [1].

Our current program target is envisaged as a demonstration of the applicability of ferritic and Fe_3Al-based ODS alloys in the high temperature heat-exchanger tubing as proposed under the proposed **DOE's Advanced Power System** program metrics, intended to sustain internal pressures (P) of up to 1000psi at service temperatures of 1000-1200°C. Within the framework of this target application, the development of suitable mechanically alloyed ferritic FeCrAl and intermetallic Fe_3Al alloy materials and processes must strive to deliver a combination of high mechanical strength at temperature and prolonged creep-life in service. Such design requirements are often at odds with each other as strengthening measures severely limit the as-processed grain size detrimental to creep life. The extrusion consolidation processes currently employed cause material flow in the longitudinal direction, resulting in extreme dispersoid and powder surface impurity fibering in the axial direction in ODS materials. Thus, elongated grains are produced aligned parallel to the longitudinal direction, with a fine grain spacing in the hoop direction. The basic problem of limited hoop creep is illustrated in Figure 2a,b within the context of the existing underlying grain structure. Fortunately ODS-alloys do exhibit intrinsic creep strength sufficient to meet design requirements albeit that this performance is only exhibited in the longitudinal direction. Ultimate failure in transverse (hoop) creep involves creep cavity concentration, Figure 2b, which strongly depends on the dominant grain boundary orientation with respect to the loading axis, Figure 4[7]. Such fibering, unless altered by post-flow forming, is expected to thwart attempts to arrive at the large transverse grain size[3,8] considered essential for improved creep performance in the hoop direction. Clearly what is required is to devise a means of effecting material flow in directions other than longitudinal that would reorient the primary fibering axis of dispersoids and impurities in the hoop direction.

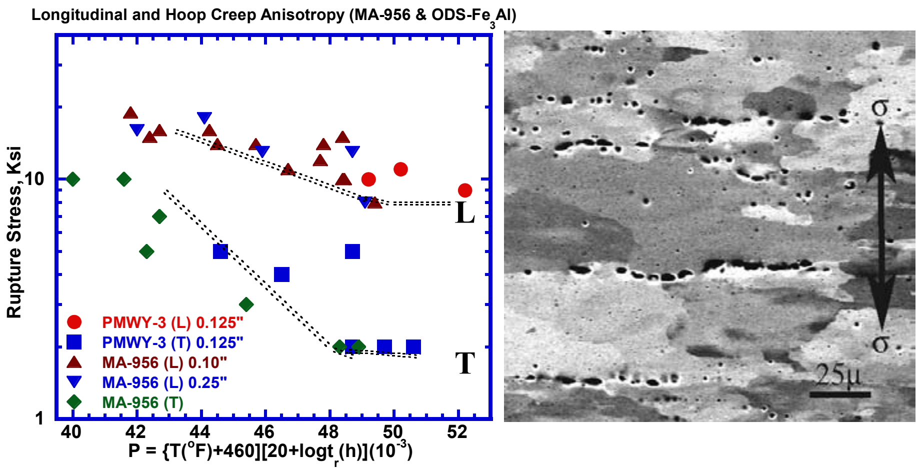

Figure 2. Longitudinal (L) vs. transverse (T) creep anisotropy in Fe_3Al (PMWY3) and MA-956 tubes. b) Creep cavitation observations in hoop creep loading tests.

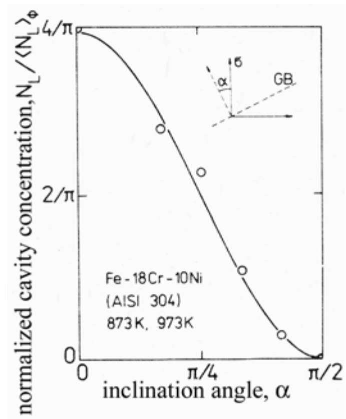

Figure 3. Creep cavity density as f_n(GB orientation) with respect to the loading axis [7].

Thus, our research objective is to modify tube-processing methodologies by incorporating cross-roll forming to create the underlying microstructure that will meet or exceed the design 'in-service' creep-life requirements of such ODS-alloy heat exchanger tubes. We are examining microscopic, microstructural and morphological issues with a view to addressing optimum material design for macroscopic components for a well prescribed 'in-service' loading criteria. This quarterly report summarizes our research activity up to the sixth quarter of performance period of January 1st 2005 - March 31st 2005. For the benefit and clarity of presentation, these reports are cumulative in order of submission.

In this quarterly performance period program work is continuing in Task 2, 3 and 4. Task 2 consisting of cross-rolling flat segments of tubes to induce grain realignment in the hoop direction. This task is continuing and engaged in iteratively. Samples of both the ODS-Fe_3Al and FeCrAl (MA956) materials have been flattened at cross-rolled. The ODS-Fe_3Al tubes are initially 1¼" OD, 1/8" wall thickness and the ODS-FeCrAl tubes are 2½" OD, ¼" wall thickness. Three different roll flattening processes are employed as 1) roll-longitudinally, 2) roll transverse to flatten and 3) roll flatten to 20-25% reduction in thickness. In Task 3, the capital equipment acquisition is complete and machinery is now undergoing installation at UCSD. The retrofit tasks of roll redesign are also proceeding concurrently. Additionally cross-rolling trials of tube samples were conducted using an outside vendor and reported here. In Task 4, samples obtained from Task 2.1 are being evaluated in transverse (hoop) creep tests. Initial results of MA956 tests reported here are compared with baseline creep performance data. The enhanced performance of cross-rolled MA956 is evident and consistent with ODS-Fe_3Al enhancements[8].

§ 2. Experimental Task Structure

The experimental work reported here is described in the context of the task structure outlined below. For the duration of this program activity through September 30th 2005 and required quarterly reporting we will refer to this task structure for clarity and precise reference.

Task 1: Extrusion Consolidations, Tube and Sheet Forms: **Completed**

1.1 ODS-Powder materials –milling studies, impurity evaluation
1.2 Annular ODS-Alloy tube and sheet extrusions

Task 2: Rolling Studies for Optimum Fibering: **Initiated & Continuing**

2.1 Single vs. cross-rolling evaluation, Parametric studies
2.2 Correlate cross-rolling strains and overall grain re-orientation

Task 3: Post-Extrusion Cross-Roll Rolling of ODS-tubes & shells: **Initiated & Continuing**

3.1 Helical/cross rolling for grain fibering
3.2 Computer model verification for torsional flow predictions

Task 4: Microstructure and Creep Performance Evaluation: **Initiated & Continuing**

4.1 Recrystallization annealing: static and gradient
4.2 Microstructure characterization & evaluation
4.3 Transverse creep and stress-rupture response

§ 3. Experimental Program Activity

Task 2.1: Single vs. cross-rolling evaluation, parametric studies: Flat sections of initial uniaxially rolled/extruded coupons to be cross-rolled *via* parametric evaluations of cross-grain fibering of the underlying grain structure.

Materials produced in Task 1.1 and 1.2 were sectioned and examined for microstructural details. No recrystallization was observed in either alloy materials as a result of this 900^{o}C thermal-mechanical treatment. This flattened strip is the required material for the initial matrix of parametric cross-rolling studies. Based on the post-forging microstructural evaluation, and in the interest of narrowing experimental windows, all further cross-rolling studies are to be conducted at 900^{o}C. Residual curvature in the forge-flattened specimens was eliminated via subsequent rolling as described here. Three separate rolling schemes were employed: 1) Rolling longitudinally in 0.01" steps till the sample was measurably flat, 2) Rolling transversely to the tube axis in 0.01" steps till the sample was measurably flat, and 3) Rolling transversely to effect a net 20-25% thickness reduction in the starting wall thickness. In the rolling schedule 3, this large deformation was accomplished in steps of 4-5% reduction per pass with the sample reheated to 900^{o}C for 15 minutes in the air furnace. The rolled flat samples are removed from their stainless steel wraps and prepared for the recrystallization treatments. Additional levels of (cross-rolling) strains are evaluated iteratively on an as needed basis. The cross-rolled specimens are recrystallized to create abnormal grain growth in such ODS-alloy coupons. The heat treatments are 1-hour at 1200^{o}C in air for ODS-Fe_3Al and a 1-hour at 1375^{o}C in air for FeCrAl (MA956). Microstructures reveal elongated grain shapes in the transverse orientation only for the sample cross-rolled 20-25% in the transverse orientation. It is likely that surface layers are affected in rolling schedule 1 and 2 but no changes are perceptible at the optical resolution level.

Task 3: Cross-Roll Rolling: As part of this task UCSD proposed to purchase and install a rotary cross-rolling set up for all cross-rolling operations of MA956 and ODS-Fe_3Al tubes under this program and any future needs. Figure 4 shows a Medart size ‘0’ straightener that was purchased, reconditioned and installed at UCSD. The size ‘0’ machine is capable of processing rod and bar in the size range of ¼” – 1½” and tubes in the size range ¼” – 2”. The machine is powered by a 30HP motor is suitable to perform cross-rolling operations at near ambient temperatures in MA956 alloys. The current roll configuration is concave-convex pair that needs to be retrofitted to convex-convex pair. Machine diagrams for this concave-convex conversion are in process. Initial efforts are on existing geometry on dummy 2” OD carbon steel tubes. By downsizing the roll diameter, we expect to cross-roll 2½” OD tube.

Figure 4 Cross-Rolling equipment currently installed at UCSD. The equipment is capable of rolling up to 1-1/2” bar/rod and up to 2-1/2” diameter alloy tubing in a continuous fashion

Sample trials to cross-roll tubes were initiated in the prior performance period employing an outside vendor. Thus ambient temperature rolling proved unsuccessful on account of equipment limitations to handle large deformation loads. As a compensating measure, MA956 tubes were preheated to 900^{o}C to reduce the overall material flow-stress for the cross rolling treatment. Single 12” section lengths of the MA956 tubes were given a total of 6-8 passes through the rolls reheating the tube after each pass for 10 minutes. Figure 5 shows the end view of a section of the tube wall after the deformation process. A straight radial (dashed red line) notch was inscribed on the end of the tube wall to monitor material flow during the deformation process. The

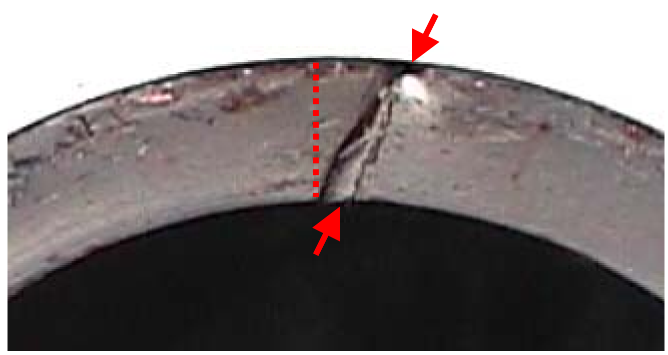

Figure 5. Cross rolling of MA956 tube via repeated passes at 900^{o}C. The red line indicates the notch as inscribed on the tube wall prior to testing. The angle between the red line and the sheared image (denoted by red arrows) gives the shear strain incurred via rotary cross-rolling process.

inclined image of the same notch (indicated by red arrows) provides a measure of the shear deformation induced by rotary cross rolling. Figure 6 shows a schematic illustration of the shear deformation process where γ indicates the actual shear strain incurred. The shear angle (γ) was measured as 35^{o} (in Figure 4) with a shear strain = 0.7. Note that the sheared notch is remarkably straight indicating that the shear deformation process is rather uniform over the entire tube wall thickness. Figure 7 (a), (b) show the before and after tube cross-section views indicating grain realignment under such rotary deformation. The as received extruded cross-section 2-dimesional views shows equi-axed structure, which undergoes a shape change under rotary deformation. The grains, with aspect ratio significantly grater than unity, are extended in the hoop direction are deemed beneficial for hoop creep performance. It is expected that ambient temperature cross-rolling may produce further grain-shape refinements.

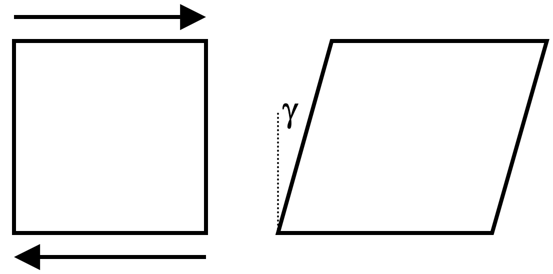

Figure 6. Schematic illustration of deformation shear strain as introduced via rotary deformation.

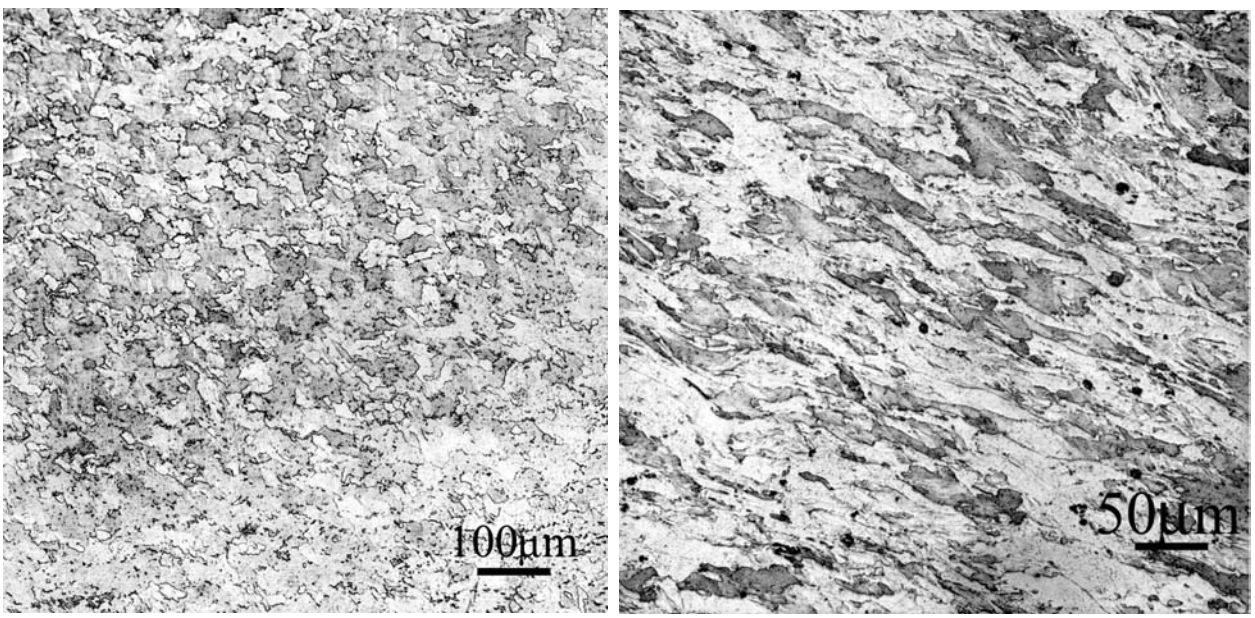

Figure 7. Cross-sectional view of the a) as –received, recrystallized and b) cross-rolled and recrystallized MA956 tube. The grain shape is significantly modified, with the grain long axis now stretched along the hoop direction.

Task 4.1: Recrystallization Annealing: Recrystallization strategies for creating abnormal grain growth in ODS alloy materials cross-rolled in Task 2.1 (and from Task 3 in later periods) suitable for transverse creep enhancements are listed in Table 1.

Table 1: Recrystallization matrix for cross-rolled ODS-Fe3Al and MA956 materials

HT Test#	Material	HT: Temperature, ^{o}C	HT: Time, Hrs	Environment
1	ODS-Fe_3Al	1200^{o}C	1 hr	Air
2	ODS-Fe_3Al	1200^{o}C	1 hr	Argon
3	ODS-MA956	1375^{o}C	1hr	Air
4	ODS-MA956	1375^{o}C	1hr	Argon
5	ODS-MA956	1400^{o}C	1hr	Air
6	ODS-MA956	1400^{o}C	1hr	Argon

The heat-treatment temperatures are based on prior DOE funded work[2-5] performed by the PI. The introduction of inert environment HT was initially applied to ODS-Fe_3Al alloys[3,8], which produced significant improvements in transverse creep. In the current performance period HT tests 1, 2, 3 and 5 have been completed for all cross-rolled materials from Task 2.1. Additional tests 4 and 6 will be conducted later in the program.

Microstructures reveal elongated grain shapes in the transverse orientation for samples cross-rolled 20-25% in the transverse orientation. It is likely that surface layers are affected in rolling schedule 1 and 2 of Task 2.1 but no changes were perceptible at the level of optical resolution.

Task 4.3: Transverse Creep and Stress Rupture Response: The initial cross-rolled samples from Task 2, heat treated in Task 4.1 are spark machined to extract ASTM E-8 standard specimens from the transverse orientation. These are being evaluated in transverse creep tests initially being performed at dead load 1-2Ksi stresses over a temperature range of 800°C – 1000°C in air.

Figure 8 shows a typical response pattern of a MA956 cross-rolled sample (cross rolled 20%) in creep tests. The sample exhibited a brief primary regime, an extended steady state regime followed by a tertiary (failure) regime. The y-axis gridline spacing denotes a creep strain of 2%. Note that the sample fails at an overall strain of about 3% with the largest component of strain (about 2%) occurring in the tertiary (failure) regime. The steady state creep rate is extracted as $9e^{-5}$/day and based on a lifetime exposure of 1381 hours the Larsen Miller parameter is computed as 48.87.

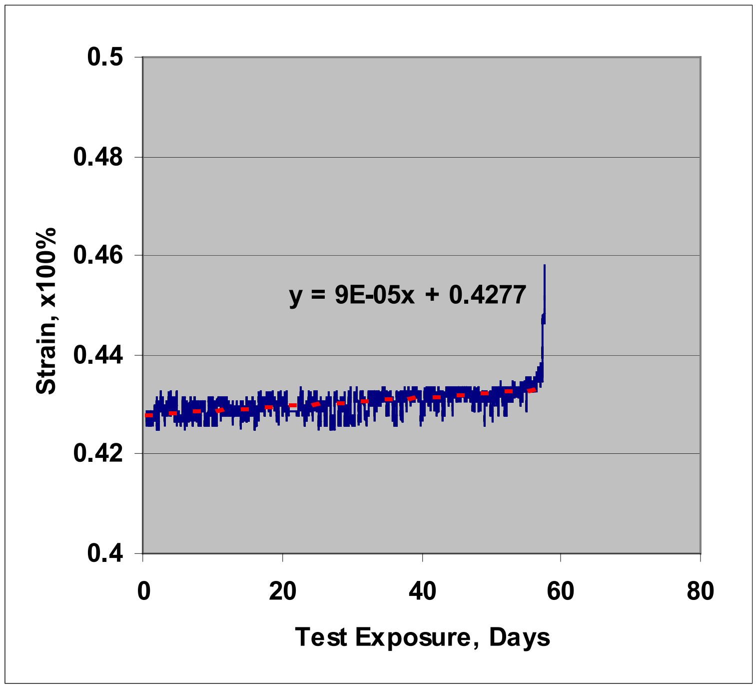

Figure 8. Creep test of a MA956 sample extracted from a 2 ½" OD, ¼" wall thickness tube and cross-rolled at 900°C to 20% reduction. Creep test conducted at 900°C at 2Ksi stress in air.

Table 2 lists all creep data compiled for MA956 base material, cross-rolled samples relevant to the current program, and flow formed specimens. Samples for the base and cross-rolled conditions were extracted from the same 2½"OD, ¼" wall thickness tube. Similar tubes are the starting material for the flow forming where the wall thickness is reduced 75-80% in several reducing steps at ambient temperatures. Tests 1-3 were run at 900°C in air at a 2Ksi stress. The mean creep rate/day (for tests 1-3) is estimated at $1.75e^{-4}$ mean creep with a mean Larsen Miller parameter = 48.67. Test 5 shows the base sample creep data with a creep rate/day of $2e^{-2}$ and a Larsen-Miller parameter value of 46.09. *Thus, at least 2 orders of magnitude improvement in creep rates is evident for the cross-rolled tests in this initial evaluation*. Finally, tests 8-10 are creep results from flow formed specimens deformed 75-80%. These test results suggest that L-M parameter is essentially similar over the 900-1000°C ranges and reconfirms the validity of accelerated creep tests. More importantly these tests indicate no further creep improvement over specimens cross-rolled only 20% (see tests 1-3 at equivalent test stress)

Table 2. Summary of creep tests performed on cross-rolled and base MA956 alloy tubes

Test	MA956 Alloy Treatment & HT	Test Temp	Test Stress	L-M Para.	rate/day
1	CR-20%@900C,HT: 1375°C-1hr,Air	900°C	2Ksi	48.87	$9e^{-5}$
2	CR-20%@900C,HT: 1375°C-1hr,Air	900°C	2Ksi	48.24	$6e^{-4}$
3	CR-20%@900C,HT: 1400°C-1hr,Air	900°C	2Ksi	48.89	$1e^{-4}$
4	CR-20%@900C,HT: 1400°C-1hr,Air	800°C	2Ksi	On-going	$6e^{-7}$
5	Flattened@900C,HT: 1375°C-1hr,Air	900°C	2Ksi	46.09	$2e^{-2}$
6	Flattened@900C,HT: 1375°C-1hr,Air	900°C	1Ksi	48.81	$2e^{-5}$
7	Flattened@900C,HT: 1375°C-1hr,Air	950°C	1Ksi	49.20	$1.5e^{-3}$
8	FlowForm@RT, HT:1375°C-1hr,Air	1000°C	2Ksi	48.77	-
9	FlowForm@RT, HT: 1375°C-1hr,Air	950°C	2ksi	48.61	-
10	FlowForm@RT, HT: 1375°C-1hr,Air	900°C	2Ksi	48.52*	-

***creep tests continuing**

We note that the ODS MA956 alloy exhibits extreme temperature and stress sensitivity as illustrated in Table 2. For example, in cross-rolled sample tests a creep rate of $1e^{-4}$ at 900°C (in test 3) drops to $6e^{-7}$ when the temperature is lowered to 800°C (in test 4). Another example is extracted from base tests 5 and 6 where the creep rate of $2e^{-2}$ at 900°C at 2Ksi drops to $2e^{-5}$ when the stress is lowered to 1Ksi. Test 7 is an extension of test 6 with the temperature raised from 900°C to 950°C that elevates the creep rate about 2 orders of magnitude from $2e^{-5}$ to $1.5e^{-3}$.

§ 4. Results and Discussion

The experimental program is proceeding at the originally prescribed timetable. The initial material preparation steps are well characterized and known from prior experience.

Significant grain alignment was recorded (Task 2.1. cross-rolling of flat MA956 samples) in the transverse direction for samples cross-rolled 20%. In the initial rotary cross rolling of MA956 tube samples we note a similar material flow in the circumferential direction as illustrated macroscopically in Figure 5 and microscopically in Figure 7. This shear deformation is nearly linear across the tube wall thickness thereby illustrating the uniformity of the rotary rolling process. The installation of an in-house rotary rolling apparatus at UCSD has been accomplished. The increased deformation capacity, on account of the larger power source, assures that materials can be rotary cross-rolled at ambient temperatures. This is beneficial in order to preserve the deformation strain energy that could produce large grains during its release upon recrystallization

Our initial intent was to compile creep data on cross-rolled samples to be compared to base data generated at ORNL. However, reference MA956 data from ORNL was compiled form samples with a different processing history. In an effort towards uniformity we are compiling our own base hoop creep data for 2 ½"OD, ¼" wall thickness tubes. In task 4, we have evaluated the performance of flat MA956 cross-rolled 20% that is deemed 1-2 orders of magnitude better than the base reference samples in creep rates. Results from flow formed materials exhibit no further enhancement suggesting that optimum effects may be achieved at lower deformation strains. The materials exhibit an extreme stress and temperature sensitivity both in the base as well as the cross-rolled condition. We expect similar results for the rotary cross-rolled tubes from Task 3.

§ 4. Conclusions

The current research program was initiated on October 1st 2003 and is concluding its sixth quarter of performance. The project progress is on schedule with work continuing in Tasks 2, 3 and 4. MA956 tubes have been successfully rotary cross-rolled while inducing significant shear strain across the tube wall thickness. This produces grain shape changes and realignment as evidenced earlier for the case of the flat cross-rolled samples.

Creep tests of cross-rolled samples illustrate their significantly enhanced creep response in terms of creep rate/day as well as overall Larsen-Miller parameter when compared to base samples. This is the original predicate of our proposed program now being realized in practice. Creep tests on rotary cross-rolled tubes are expected to show similar improvements as well. Initial test results on flow formed MA956 (reduced 75-80% in wall-thickness) indicate that the creep performance is about the same as for samples cross-rolled 20%. This suggests that the beneficial effects of grain shape control are perhaps achieved at lower levels of deformation strain. These issues will be fully explored in the remainder of this program.

References:

1. F. Starr, in *Materials for High-Temperature Power Generation and Process Plant Applications*, A. Strang, Ed., IOM Communications Ltd., Book No. 728, pp. 79-152 (2000).
2. V.K. Sikka, I.G. Wright, and B.K. Kad, " Processing of Oxide-Dispersion-Strengthened Fe_3Al-Based Alloy Tube" (1998) *12th Annual Fossil Energy Materials Conf.*, Knoxville, TN, May 12-14, 1998, p.11-19
3. B.K. Kad, C.G. McKamey, I.G. Wright and V.K. Sikka, "Optimization of ODS-Alloy Properties" (2002) *16th Annual Fossil Energy Materials Conf.*, Baltimore, MD, April 22nd –24th, 2002.
4. B.K. Kad, C.G. McKamey, I.G. Wright and V.K. Sikka, "High Temperature Mechanical Properties of ODS-Fe_3Al Tubes" (2001) *15th Annual Fossil Energy Materials Conf.*, Knoxville, TN, April 30th -May 2nd, 2001
5. B.K. Kad, V.K. Sikka, R.N. Wright and I.G. Wright, "Oxide Dispersion Strengthened Fe_3Al-Based Alloy Tubes" (2000) *14th Annual Fossil Energy Materials Conf.*, Knoxville, TN, April 24-26, 2000; *ibid* (1999) *13th Annual Fossil Energy Materials Conf.*, Knoxville, TN, May 11th –14t, 1999.
6. J. Ritherdon, A.R. Jones and I.G. Wright, “The Recovery and Recrystallization of a Mechanically Alloyed ODS-Fe_3Al Alloy” (2001) *Materials Science Forum*, **360-362**, pp.217-222, 2001
7. I.W. Chen and A.S. Argon, “Creep Cavitation in 304 Stainless Steel” (1981) *Acta Metallurgica*, **29**, p.1321, 1981; *ibid.* “Diffusive Growth of Grain-Boundary Cavities” *Acta Metallurgica*, **29**, p1759, 1981
8. B.K. Kad, I.G. Wright, V.K. Sikka and R.R. Judkins, (2004) “Optimization of Hoop Creep Response in ODS-Fe_3Al Tubes” *18th Annual Fossil Energy Materials Conf.*, Knoxville, TN, June 2nd – 4th, 2004