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Dimensional Stability of Superinvar

Steven R. Patterson

Lawrence Livermore National Laboratory
P.O. Box 808, L-332, Livermore, California 94550

ABSTRACT

The unloaded dimensional stability and thermal expansivity of a single furnace melt of superinvar have been measured at room temperature using interferometric techniques. Thermal expansivity has been determined with an uncertainty of several parts in 10^8 per degree centigrade, while dimensional stability has been determined with an uncertainty of order one part in 10^9 per day. Samples subjected to plastic deformation in their processing history displayed a stability improvement from 20.5 x 10^{-6}/day to 5.5 x 10^{-9}/day and a reduction in thermal expansivity from 0.56 x 10^{-6}/°C to 0.23 x 10^{-6}/°C associated with the increased mechanical work in the material.

1. MATERIAL SELECTION AND PROCESSING

Modern scientific instruments place increasingly stringent demands on the dimensional stability of materials used for the construction of precision structures. The dimensional change of a material due to a change in temperature is characterized by its coefficient of thermal expansion (CTE). Dimensional changes may also occur over time in a fixed environment. In considering the selection of a material for demanding applications, both the CTE and temporal stability must be considered. Indeed, with reasonable control of the ambient temperature, pressure, humidity and magnetic field, temporal instability may be the factor limiting performance.

Superinvar, the material studied here, is one alloy in a class of materials of practical interest for the construction of precision systems. The invars are relatively inexpensive materials (by comparison with other low-CTE materials) and may be fabricated using conventional metalworking techniques. Superinvar is of particular interest because it exhibits a lower CTE than the other invars, and because of an early report by Jacobs, Bradford and Berthold indicating that it possessed a particularly high degree of temporal stability.[1] This paper reports a series of CTE and temporal stability measurements performed on superinvar samples taken from the same melt but subjected to different degrees of processing.

One of the experimental aims of this study was to learn about the conditions leading to the single high stability result reported by Jacobs. Accordingly, the chemistry of the material was specified as a high purity superinvar with 31% Ni and 5% Co. This alloy lies at the minimum CTE point of the Fe-Ni-Co system[2], and is the nominal alloy used by Jacobs. Table I shows the composition which was specified, the analysis performed at the mill at the time of melt and an independent analysis of the material after receipt at Livermore. For reference purposes, the composition of the sample measured by Jacobs is also listed.[3]

The material used in the samples was cast in an air induction furnace as part of a large order. Although the same steel mill produced both this material and Jacobs' samples, the furnaces used for the two melts were different: Jacobs' sample came from a melt in a smaller furnace. While it may be argued that a smaller melt allows better control of the furnace and load, the only published study[4] relating melt size to physical properties of superinvar demonstrates that a larger melt is superior. It is reasonable to assume, then, that some factor other than the melt size is responsible for the high stability of the small melt sample.

To assess the effect of processing on the CTE and stability of the samples, stability samples were cut from the original ingot (INGOT), and intermediate forged billet (RECT) and finished cross-rolled plate (RS). The initial forging temperature was 1177°C to 1191°C (2150°F to 2175°F). Forging was completed at a temperature of 982°C (1800°F).

Rolling was started with the temperature at 1177°C (2150°F) and completed with the temperature in the range of 871°C to 927°C (1600°F to 1700°F). These temperatures are sufficiently below those required for full recrystallization so that significant reduction of grain size occurred during the processing.

Cylindrical samples 38mm in diameter and 100mm long were fabricated using normal machine shop practices. The finish cut on the cylindrical diameters was taken with a freshly sharpened carbide tool at a 0.025 mm depth of cut to minimize modification of the sample. No change in microstructure could be observed on the diameters. The two ends of the stability samples were polished using standard optical shop techniques.

In addition to these samples, one sample (A) was constructed from 38 mm square metal bars of rolled material using the weld sequence illustrated in Fig. 1. . Half of the sample so produced consists of weld filler metal. The use of alternate welds on opposite sides of the sample results in a worst-case application of thermomechanical stress to the sample. Pure superinvar from the same melt was used for the weld filler material in this sample. This welded sample displayed severe cracking throughout the weld region.

Table I Chemical composition of superinvar (all values in weight percent)

Element	Specification	Mill Analysis	LLNL Analysis	Jacobs' Sample
Ni	31-32	31.33	31.6	31.2
Co	5.0-5.5	5.46	5.49	5.10
Mn	0.35-0.45	0.39	0.39	
C	<0.05	0.030	0.016	0.03
Al	<0.10		0.13	<0.01
Ca			<0.01	
Pb	<0.005		<0.01	
N	<0.01		0.003	
P	<0.015	0.012	0.009	0.005
S	<0.015	0.005	0.005	0.013
Si	<0.15	0.15	0.16	0.06
Ti			<0.005	<0.01
O	<0.01		0.005	

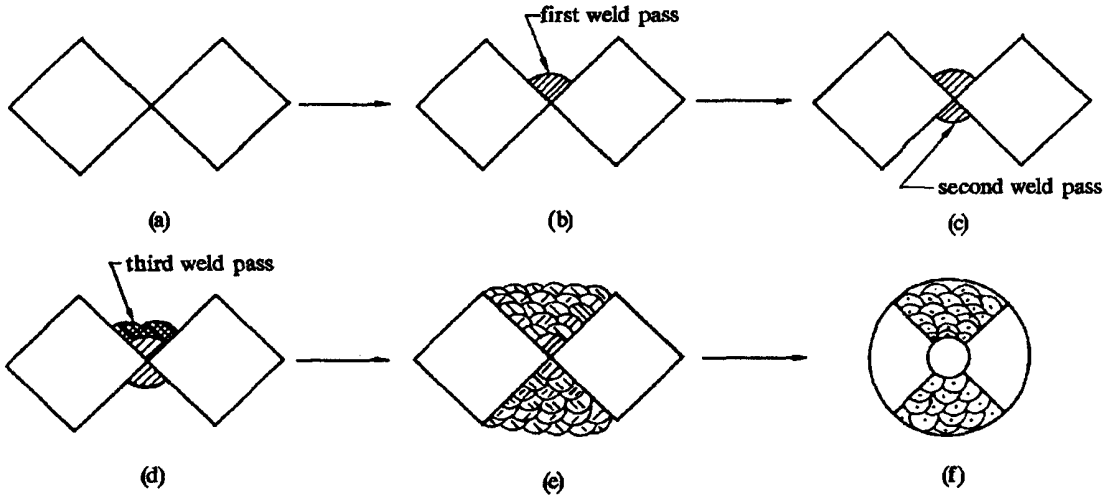

Fig. 1. Construction of welded test samples (cross section): (a) starting base metal bars, (b) first welding pass, (c) second welding pass, (d) third welding pass, (e) finished weld, (f) final test sample.

All samples were heat-treated according to a schedule derived from that developed by Lement, Averbach and Cohen[5] for the optimum combination of low CTE and high stability of invar. This treatment has become something of a de facto standard for the treatment of superinvar as well as invar despite the lack of quantitative studies of heat treatment effects in superinvar. This treatment consists of heating the samples to 830°C for 20 minutes, then rapidly (<10 sec) quenching to less than 35°C in water. This is followed by one hour of stress relieving at 315°C in an inert atmosphere. Finally, the

sample is heated to 95°C and held at that temperature for 48 hours. The homogeneous samples were rough machined before the quenching step, and finished machined before stress relief. End polishing was accomplished after all heat treating steps. The welded sample was fabricated completely between the quench and stress relief steps of the heat treatment.

All samples were polished, etched and examined under magnification to determine grain structure. With the exception of the ingot specimen, all of the samples displayed equiaxe grains of austenite with no evidence of martensite. The ingot specimen displayed very large grains with aspect ratios in the range of 3:1 to 10:1. Twinning could be discerned in all of the micrographs. The weld metal region of sample A displayed a regular network of segregation inside large grains. The weld metal region of sample A also showed severe intergranular cracking.

Table II Hardness and grain size

Sample	Average grain size (microns)	Average hardness (Vickers)
INGOT	383	146.6 ± 4.0
RECT	42	143.6 ± 1.0
RS	38	142.4 ± 7.4
A (base)	72	151.8 ± 5.8
A (weld)	350	150.6 ± 4.6

To calculate grain size, a set of grains was selected from a random array of points on each metallograph. The square root of the average area of the grains in selected set is taken as the average grain size of the material. This method effectively weights the grain size distribution by the volume fraction of each size of grain. Microhardness of samples was measured in accordance with ASTM Standard E-384-73. The result of grain size and microhardness measurements is shown in Table II. The quoted error for microhardness is the standard deviation of measurements at five locations on each sample. This error is significantly larger than systematic errors in the measurement itself.

2. MEASUREMENT METHOD

Stability and CTE measurements were performed using a precision dilatometer of design similar to that employed by Berthold, Jacobs and Norton.[6,7] The system measures the strain in a sample by tracking the transmission maximum of a spherical confocal Fabry-Perot interferometer constructed using the sample as a spacer between the two mirrors. This is accomplished by polishing the ends of the sample and optically contacting a mirror to each. Because of the use of a spherical confocal configuration, there is some tolerance of both absolute length variation and parallelism errors between the end faces of the sample.

The system is illustrated schematically in Fig. 2. . A HeNe laser with adjustable cavity length (Spectra-Physics model 119) illuminates and is wavelength locked to the transmission maximum of the Fabry-Perot cavity containing the sample spacer. The frequency of this laser is determined by heterodyne detection of a sample of its beam using an atomic line stabilized laser as a local oscillator. Continuous monitoring of the heterodyne frequency provides a direct measure of the spacer strain. To prevent feedback from the Fabry-Perot cavity from perturbing the illuminating laser cavity, a polarization isolator is used. This isolator consists of a polarizer and quarter wave phase retarding plate whose axis is rotated 45° from that of the polarizer so that the radiation reaching the Fabry-Perot cavity is circularly polarized. Reflection from the Fabry-Perot interferometer reverses the sense of polarization of the beam, which is subsequently converted to a linearly polarized beam by the quarter wave plate and is rejected by the polarizer. Such an isolator has been shown to be effective in isolating spherical confocal Fabry-Perot interferometers of very high finesse.[8]

The dilatometer was designed to measure temporal stability with a resolution of less than 5×10^{-9}/day in a test time of less than two weeks. To accomplish this requirement, the measurement is carried out in an evacuated chamber which is temperature controlled with a stability of 0.003°C. The Fabry-Perot cavity is initially aligned and subsequently monitored for alignment using the mode matching technique described by Munnerlyn and Balliett.[9] Locking of the illuminating laser is accomplished by modulating the laser frequency cavity length slightly at a 5 KHz rate. A phase-sensitive detector connected to the photodiode at the output of the Fabry-Perot cavity is then used to drive the average length of the laser cavity to eliminate the first derivative of the modulation in the interferometer transmission.

When all errors are considered, the overall estimated strain measurement error of the dilatometer is 10^{-8} with an additional drift of 10^{-10}/day. To measure CTE, the temperature of the chamber is changed by ± 0.56 °C and the resulting strain measured. This technique provides a measure of CTE with a peak error of approximately 3 x 10^{-8}/°C.

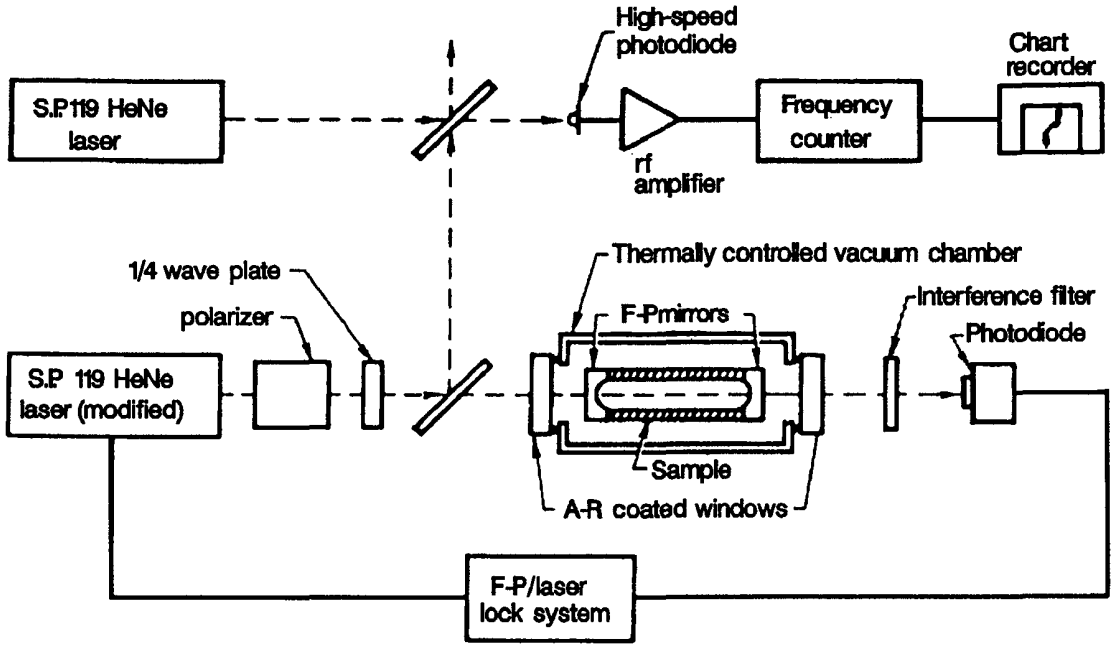

Fig. 2. Overall design of the Fabry-Perot dilatometer.

3. MEASURED STABILITY AND COEFFICIENT OF THERMAL EXPANSION

Each sample was tested for a period of several hundred to a thousand hours. The observed strain as a function time is shown in Fig. 3. through Fig. 6. . Short sections of straight lines indicate periods during which isothermal strain data was not available either because the chamber temperature was changed to measure the coefficient of thermal expansion, or because of equipment failure not affecting the alignment of the lasers and the interferometer.

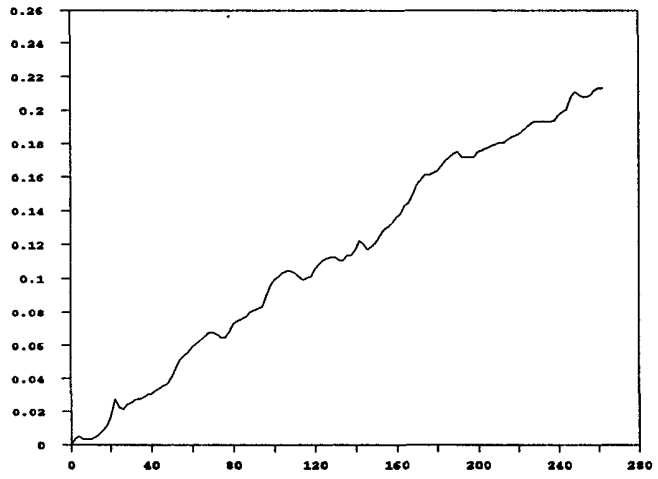

Fig. 3. INGOT sample microstrain vs. time (hours)

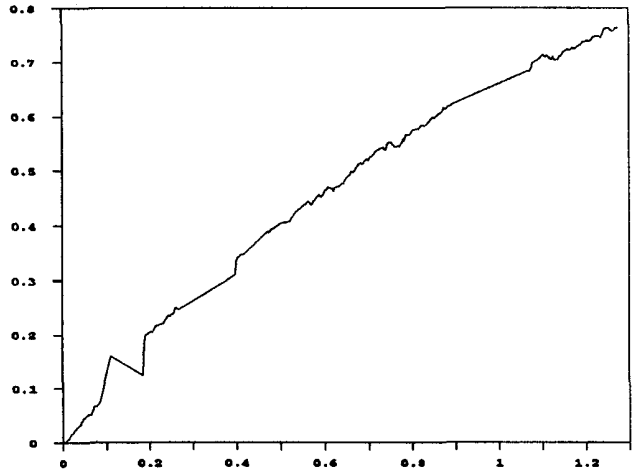

Fig. 4. Sample RECT microstrain vs. time (K hours)

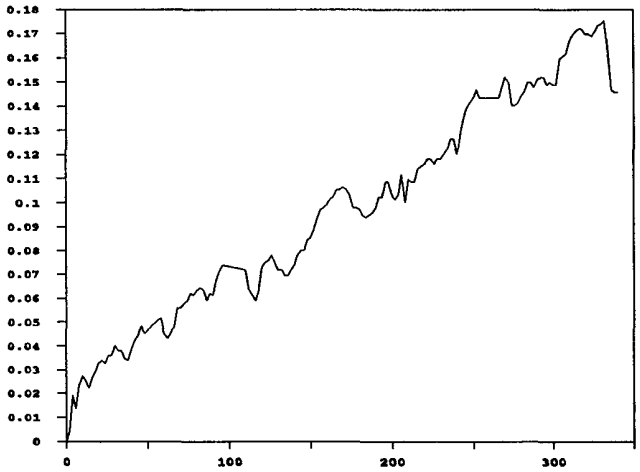

Fig. 5. Sample RS microstrain vs. time (hours)

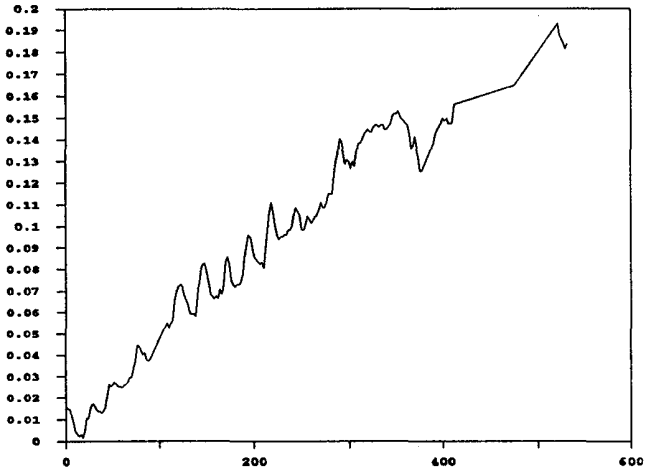

Fig. 6. Sample A microstrain vs. time (hours)

All of the curves are consistent with a smooth and continuous dilatation of the samples. The short-term noise that is present on these curves is consistent with fluctuations in laboratory temperature. While the observed instability may arise from the relief of residual stresses in the samples or from slow thermally activated structural transformation in the material, residual stress relief is a less likely explanation given the consistent expansion observed in all of the samples. Moreover, there is no analogous stress relief behavior reported in binary invar alloys. Lement, Averbach and Cohen have observed residual stress relief in cylindrical samples of invar subjected to the same heat treatment used here. In their experiments, stress relief was characterized by contraction along the length of the cylinder. Candidates for thermally activated transformations include phase transformations in the bulk material either between forms of austenite or between austenite and martensite, redistribution of alloying materials and diffusion to or from grain boundaries.

The longest of the stability measurements (RECT) shows a statistically significant deviation from linear drift. The data for RECT may be fitted to a curve of the form

$$\epsilon = 1.126\times10^{-6}\ (1-e^{-t/1150hr}) \qquad (1)$$

with an rms error of 7.6×10^{-9}, well within experimental errors. A similar exponential form with a time constant of 1150 hours provides an improved fit relative to a straight line for all of the other stability data, but the improvement is not statistically significant in light of the experimental errors present.

The measured values of CTE for samples INGOT, RECT, RS and A were 0.56×10^{-6}/°C, 0.45×10^{-6}/°C, 0.34×10^{-6}/°C and 0.23×10^{-6}/°C, respectively. These values are plotted against the measured temporal stability of each sample in Fig. 7. The data corresponding to the high stability reported by Berthold, Jacobs and Norton. This sample has since been considered to display anomalously low CTE and high stability.[10] In the light of the present data, however, this point appears to represent a consistent trend in material properties due to metallurgical history and/or condition independent of the composition and heat treatment.

The data for the homogeneous samples INGOT, RECT and RS suggest that the improved stability and lower CTE are associated with increasing amounts of mechanical work in the material. The weld sample A is also consistent with this conjecture, with the work being supplied by the differential expansion of the material during successive welding and cooling on alternate sides of the sample during fabrication.

Mechanical work may lead to these effects through one of several influences on the material: (1) reduction of grain size, (2) introduction of lattice strain, (3) increase in the dislocation density, or (4) improved mixing of impurities. The improved stability and reduced CTE of sample A are inconsistent with a dependence on grain size; moreover, sample A also shows a high degree of segregation in the photomicrographs which makes it unlikely that it has a greater degree

of impurity homogenization.

An increase in dislocation density should be accompanied by a measurable work hardening. The series of samples INGOT to RS display successive improvements in thermal and stability properties, but no increase in harness. This suggests that either the working temperature or the temperature of the first heat treatment was sufficiently high to allow recovery of the crystal structure within the grains of the material. In the case of the welded sample, welding was followed only by the lower temperature heat treatment steps, so that work hardening is evident.

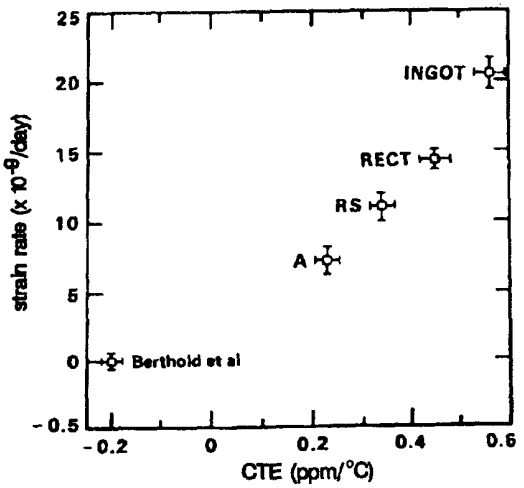

Fig. 7. Temporal stability vs. CTE for several samples of superinvar.

The most attractive potential mechanism for the improvement of stability and reduction of CTE is the introduction of lattice strain during mechanical work. While such strain may be reduced by exposure of the samples to temperatures that allow recovery of dislocations, it will not be fully relieved until the temperature is raised above the recrystallization temperature. If the influence of such strain is to increase the magnitude of the temperature dependence of spontaneous magnetostriction or saturation magnetization in the material, an associated lowering of the CTE would be expected.

4. CONCLUSIONS

Measurements of the temporal stability and coefficient of thermal expansion of superinvar with low levels of impurities have been made with resolutions of order 10^{-9}/day and 10^{-8}/°C respectively. These measurements show that both the stability and CTE are systematic functions of the amount of mechanical work to which the material is subjected during its processing history. The results suggest that the very high stability and slightly negative room temperature CTE reported earlier for superinvar may be consistent with a high degree of mechanical work in presently available melts of the material.

Although the data presented here are very suggestive of a smooth connection to this earlier data, there remains a substantial gap between those measurements and these which can only be bridged by extrapolation at the present time. This suggests the desirability of an additional series of experiments using more heavily worked materials.

5. ACKNOWLEDGEMENTS

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