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AN INVESTIGATION OF W-3%RE AND W-25%RE THERMOELEMENTS IN VACUUM, ARGON AND HYDROGEN

by

G. W. Burns and W. S. Hurst

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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U.S. DEPARTMENT OF COMMERCE
National Bureau of Standards
INSTITUTE FOR BASIC STANDARDS
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FINAL REPORT

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Technical Management
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U.S. DEPARTMENT OF COMMERCE
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FOREWORD

The work described herein was performed at the National Bureau of Standards (Temperature Section, Heat Division, Institute for Basic Standards) under Interagency Agreement NASA Order No. C-30968-B with Mr. Peter Cipollone of NASA Lewis Research Center as Technical Project Manager, and Mr. George Glawe of NASA Lewis Research Center as Technical Consultant.

ABSTRACT

The effect of exposure of bare wire W-3% Re and W-25% Re thermoelements to environments of high vacuum ($< 1 \times 10^{-8}$ torr), Argon and Hydrogen at temperatures ranging from 2200 K to 2600 K for periods up to 1000 hours has been investigated. The individual thermoelements, and hence the W-3% Re versus W-25% Re thermocouple pair, experienced a shift in the emf-temperature relationship on initial heating. In Argon and Hydrogen, the shift was less than 1% at 2000 K for the thermocouple pair. After the initial shift, the thermoelements exposed in Argon and Hydrogen experienced no significant further change in their emf-temperature relationship for periods up to 1000 hours. Both the W-3% Re and the W-25% Re thermoelements, when exposed in vacuum at 2400 K and above, drifted continually from original calibration as a result of preferential loss of Re by evaporation. The thermoelements were examined for chemical and structural changes by electron microprobe analysis and by conventional metallographic techniques.

TABLE OF CONTENTS

AN INVESTIGATION OF W-3% RE AND W-25% RE THERMOELEMENTS IN VACUUM, ARGON AND HYDROGEN

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National Bureau of Standards
Temperature Section

SUMMARY

This reports an investigation of the effect of the exposure of bare wire (0.25 mm diameter) W-3% Re and W-25% Re thermoelements to a temperature of 2400 K for periods up to 1000 hours. Environments for the tests consisted of vacuum, high purity Argon, or high purity Hydrogen. Some tests at 2200 K and 2600 K in vacuum were also performed. The thermocouple wires were thermally exposed by electrically self-heating the wires within a bakeable stainless steel test chamber in which either high vacuum ($< 1 \times 10^{-8}$ torr) or high purity gaseous atmospheres were created. The change in calibration of the thermoelements after thermal exposure was determined in a high temperature calibration furnace. The changes were found by comparison of the exposed thermoelements against previously unheated thermoelements from the same wire lots.

The thermoelements, and hence the W-3% Re versus W-25% Re thermocouples, exhibited a shift in their emf-temperature relationships after aging above 2200 K in all three environments. The aging period necessary to complete the shift at 2400 K is less than 1 hour. The magnitude of the shift varied with the wire lot. After thermal exposure of 50 hours in Argon at 2400 K, the shift in calibration at 2073 K (1800 °C) of W-3% Re versus W-25% Re thermocouples from three different matched wire lots was +17 K, +3 K, and +6 K. There was also a dependence of the shift magnitude upon the type of environment: for one matched lot of wire, the shift in the calibration of the thermocouples at 2073 K after thermal exposure at 2400 K for 50 hours was +17 K in Argon and +9 K in Hydrogen.

After the initial shift, the thermocouples exposed at 2400 K in high purity Argon and Hydrogen experienced no further significant change in calibration for time periods up to 1000 hours. The thermocouples exposed to temperatures higher than 2200 K in high vacuum ($\leq 1 \times 10^{-8}$ torr) drifted in calibration as a result of the preferential loss of Re from the W-Re alloys. The drift in calibration was strongly dependent upon both the exposure temperature and time. At 2073 K (1800 °C) a drift in the thermocouple calibration of -175 K after 500 hours exposure at 2400 K was typical, while a drift of about the same magnitude occurred only after 50 hours exposure at 2600 K. The drift in calibration after exposure for 500 hours at 2200 K was less than 5 K.

Metallographic studies and qualitative tests of bend ductility were performed in order to more fully characterize the exposed thermoelements. The W-25% Re thermoelements experienced large equiaxial grain growth with thermal exposure and became quite fragile, retaining only very limited room temperature ductility. The W-3% Re thermoelement, which is a "doped" alloy, remained quite ductile after thermal exposure, especially in the case of the Hydrogen and vacuum environments. The onset of secondary recrystallization was observed for W-3% Re thermoelements exposed in vacuum and Hydrogen, with the wire developing a central core of very large interlocking elongated grains, while only primary recrystallization (broadening of the fibrous grain structures) was observed for the thermoelements exposed in Argon. The occurrence of secondary recrystallization (appearance of central core) did not appear to strongly influence the magnitude of the shift in thermal emf.

INTRODUCTION

Recent advances in the technology of propulsion and space power systems have increased demands on temperature measuring systems that must operate at high temperatures in various environments for long periods of time. While W-Re alloy thermocouples have been used as high temperature sensing devices, reliable operation has not always been achieved, especially when long term use, (e.g., thousands of hours) is required at elevated temperatures. Most previous studies have been performed without highly controlled characterization of materials and environments, but the presence of small amounts of impurities has the potential of greatly influencing thermocouple performance (Ref. 1). Therefore, tests in more carefully controlled conditions were desirable. An examination of carefully characterized materials in controlled environments could aid in the identification of those parameters having gross effect on the W-Re thermocouple performance.

Towards achieving this, these studies were performed with bare W-Re thermocouple wire in well characterized environments. The results obtained give pertinent information which defines some of the limiting capabilities of tungsten-rhenium thermocouples as high temperature measuring instruments. It should be recognized that the addition of insulators and sheaths, as most users of thermocouples require, introduces additional variables, such as problems in chemical compatability of the materials, and that most previous tests have been performed with insulators and sheaths. The performance of the bare wire gives a basis against which the performance of the inherently more complicated insulated and sheathed thermocouples may be compared, both as regards to the thermoelectric drift and as regards to the chemical and metallurgical behavior of the materials.

Accordingly, this work involves an investigation of the changes in the thermal emf of bare wire W-nominally 3% Re versus W-nominally 25% Re thermocouples after exposure to temperatures up to 2600 K in environments of vacuum ($< 1 \times 10^{-8}$ torr) or Hydrogen gas (nominally 1 atm.) or Argon gas (nominally 1 atm.) for periods up to 1000 hours. Analyses were performed on the chemical composition of both the wire alloys and the gases, and on the changes in the metallurgical microstructure that developed during the tests.

EXPERIMENTAL METHODS

RATIONALE FOR METHOD OF TESTING

Let us assume that the thermoelements and consequently the thermocouple undergo changes in calibration as a result of thermally activated processes which require a temperature above ambient room temperature in order to occur (for example, relief of internal stresses). Further, let us assume that such processes will be completed within some finite time, depending upon that temperature, after which the thermocouple becomes thermally aged and will undergo no further change in calibration. (Other processes could result in the continual change of the thermocouple calibration, of course, such as chemical changes in the materials resulting from either contamination, or from the loss of impurities or of a major constituent by vaporization).

If such a thermally unaged thermocouple is exposed to high temperature in such a manner (as in a calibration furnace) that the temperature along each thermoelement varies from a maximum temperature at the measuring junction to ambient temperature outside the furnace, then only part of the length of each thermoelement is subjected to

temperatures sufficiently high for thermal aging to occur, and a transition from thermally aged to unaged material will exist along each thermoelement. In general, the magnitude of the change or shift in calibration of a thermocouple exposed and thermally aged in this manner would be less than the magnitude of the change or shift in calibration of a thermocouple whose thermoelements have been uniformly thermally aged along their entire lengths. Therefore, in these studies, in order to "magnify" the change in thermocouple calibration, the thermocouple materials were uniformly thermally exposed over their entire lengths. (It follows that, if "as received" bare wire thermocouples were to be used in typical furnace applications, the shift and/or drift in calibration obtained would be less than the values given in this report, for a given lot of wire).

In instrumentation with a thermocouple where only part of the "as received" (unaged) W-3% Re and W-25% Re thermoelements are subjected to high temperature, the user should be aware that the calibration will no longer be unique, but will depend to a certain extent on the temperature distribution along the thermoelements during subsequent use. This problem could be essentially eliminated by uniformly aging the thermoelements before use. However, if this were not possible, the thermocouple should be used at constant immersion. This will in many applications result in a fairly reproducible temperature distribution along the thermoelements. Under such conditions the calibration of the thermocouple would be fairly reproducible.

TEST APPARATUS AND PROCEDURES

In preparation for thermal exposure, a loop of a bare thermocouple wire (90 to 100 cm long) was hung vertically from two electrical feedthroughs within a bakeable stainless steel test chamber (Fig. 1). The test chamber was then evacuated, using only molecular sieve sorption roughing pumps and high vacuum ion pumps, in order to eliminate the possibility of hydrocarbon backstreaming which could be present in an inadequately trapped diffusion pump system. Vacuum levels of 5×10^{-9} torr* and lower were achieved routinely with these systems within 24 hours after mild bakeouts at approximately 400 K. Static gas environments of Argon or Hydrogen were created by back-filling the evacuated test chamber to 1 atmosphere pressure from a cylinder of high purity compressed gas through a stainless steel manifold. For the tests in vacuum, the pressure was maintained with a 200 liter/second ion pump at less than 1×10^{-8} torr with the wires at test temperature.

*All pressures are indicated equivalent nitrogen pressures.

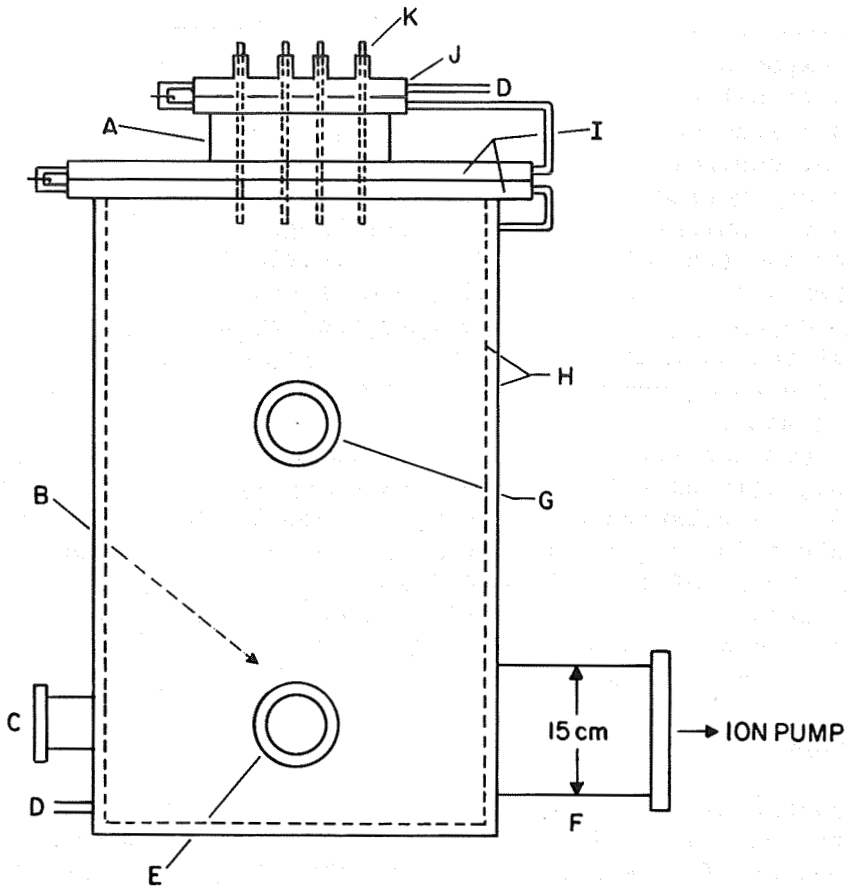

Fig. 1. Typical test chamber for thermal exposure of thermoelements: (A) 15 cm (6") Type 304 stainless steel tube; (B) Bayard-Alpert type ionization gauge on port in rear; (C) To 1 cm (3/8" dia.) stainless steel gas manifold, with all-stainless steel valve; (D) To cooling water; (E) 2.5 cm (1") stainless steel ultra-high vacuum valve with Au seat; (F) 15 cm (6") Type 304 stainless steel tubulation, equipped with ultra-high vacuum flange; (G) 3.8 cm (1-1/2") Corning 7056 glass viewing port on ultra-high vacuum stainless steel flange; (H) Double wall, Type 304 stainless steel, 25 cm (10") diameter; (I) 30 cm (12") o.d. stainless steel flange, gold O-Ring seal; (J) 20 cm (8") o.d. stainless steel flange, Cu gasket seal; (K) Ceramic insulated electrical feedthroughs. Typically, 12 to 16 provided.

From two to six wire loops were mounted in the chamber and thermally exposed simultaneously by electrically self-heating with 60 Hertz alternating current. The heating current to each wire loop was adjusted separately by manual regulation of a solid-state A.C. current controller. The temperature of each wire loop was determined and monitored during the thermal exposure with a calibrated visual optical pyrometer. Corrections were applied to account for the transmittance of the glass viewing window on the test chamber and the spectral emittance of the test wires. By these procedures, it is estimated that the temperature of the wires were set at the thermal exposure temperatures of 2200, 2400 and 2600 K within an accuracy of ±50 K. However, the procedure for setting and monitoring the temperature with the same optical pyrometer, coupled with careful monitoring of the heating currents during test, allowed all wires (of one alloy type) to be set and controlled on a relative basis to within about ±25 K. All values of temperature in this report are given on the International Practical Temperature Scale of 1948 (IPTS-48) in degrees Kelvin (in some instances equivalent values are also given in degrees Celsius). Thermodynamic temperatures may be obtained, if desired, by conversion of values of temperature on the IPTS-48 to the values of temperature on the recently adopted IPTS-68, which closely represents the thermodynamic temperature (Ref. 2).

The changes in the thermal emf of W-3% Re and W-25% Re thermocouple wires were determined after heating (thermal exposure) at high temperature for periods of 1, 50, 100, 150, 250, 500, 750 and 1000 hours in the environments of Argon, Hydrogen, or vacuum. The calibrations of the thermally exposed wires were performed in a high temperature resistance heated furnace (Ref. 3) with an Argon environment by determining the emf of the thermally exposed (test) wires versus "as received" (reference) wires [from the same spool (lot) as the exposed wires] at 200 degree intervals between 673 and 2073 K (400° and 1800 °C).

For these calibrations, a common measuring junction was formed between thermally exposed W-3% Re and W-25% Re test wires and W-3% Re and W-25% Re reference wires by welding with a D.C. arc in helium. The common measuring junction was heated in the calibration furnace and the emf of the W-3% Re test wire versus the W-3% Re reference wire and of the W-25% Re test wire versus the W-25% Re reference wire was measured with a precision potentiometer having a resolution of 0.1 microvolt and a limit of error of not more than ±(0.015% of reading +0.5 microvolt). The temperature of the common measuring junction was determined with the thermocouple formed by the W-3% Re and W-25% Re "as received" reference wires to within an

estimated accuracy of ±0.5% at each calibration point. For all calibrations the temperature of reference junctions was maintained at 273.15 ±0.03 K (0 ±0.03 °C) with an ice bath.

The calibration data gives the changes (with respect to the "as received" reference wires) in thermal emf, due to exposure, of each thermoelement. The change in the thermal emf of a W-3% Re versus W-25% Re thermocouple, due to exposure, is then obtained by combining the changes measured for the separately exposed thermoelements; that is given by the algebraic difference between the change in emf of the W-3% Re thermoelement and the change in emf of the W-25% Re thermoelement.

This procedure for determining the change in the thermal emf of the test wires is a conventional one. It eliminates the need for extreme accuracy in the measurement of the temperature level since the thermoelectric power (thermal emf per degree) of the test wires versus reference wires of nominally the same type is small. The procedure does require that the test wire lots have reasonably good thermoelectric uniformity, so that samples (reference or test) taken at random from the wire lot will be very nearly identical thermoelectrically.

The estimated maximum uncertainty in the measurement of the changes in thermal emf at 1273 K (1000 °C) and 2073 K (1800 °C) for both the elements and thermocouples is tabulated in Table I. The major source of error is the inhomogeneity of the wire lots. The small differences in the estimated uncertainties between the gaseous and vacuum environments, however, results primarily from differences occurring in the thermoelectric power (of the exposed test wires versus the "as received" reference wires) in the two cases.

It was recognized in obtaining the calibration data that the "as received" reference wire is subjected to some aging during the calibration process. In the case of the W-25% Re element, a very slight effect of aging could be observed at temperatures of 1873 K and above. However, the time at the higher temperatures was short so that the effect on the calibration data is small and is of little consequence, since to a first approximation the effect is the same in each calibration.

The thermoelectric changes for the thermocouples are also expressed in terms of the equivalent temperature change (ΔT). For the thermocouple, ΔT is obtained by dividing the change in the thermal emf of the thermocouple at a given temperature by the thermoelectric power (dE/dT) at that temperature. ΔT for the separate thermoelements

TABLE I

Estimated maximum uncertainty in the measurement of the change in the thermal emf of exposed thermoelements and thermocouples.

Exposed in Argon and H_2 at 2400 K or in Vacuum at 2200 K

Calibration Temperature	W-3% Re Thermoelements	W-25% Re Thermoelements	W-3% Re vs W-25% Re Thermocouples	
				(equiv)
Degrees K	μv	μv	μv	K
1273	±20	±20	±40	±2.0
2073	±30	±30	±60	±3.8

Exposed in Vacuum at 2400 and 2600 K

Calibration Temperature	W-3% Re Thermoelements	W-25% Re Thermoelements	W-3% Re vs W-25% Re Thermocouples	
				(equiv)
Degrees K	μv	μv	μv	K
1273	±25	±30	±55	±2.8
2073	±35	±50	±85	±5.4

is obtained similarly. However, for the W-25% Re thermoelement (negative element of the thermocouple), an additional multiplicative factor of (-1) is included, so that a positive change in emf results in a negative ΔT. In this manner, the effect of thermal exposure of the separate thermoelements is expressed in terms of its temperature effect upon the thermocouple, and the algebraic sum of the ΔT's for each of the thermoelements equals the ΔT for the thermocouple.

MATERIALS TESTED

The studies were conducted using three different matched lots of commercially available thermocouple wire, designated W-3% Re and W-25% Re. The thermocouple wires of each lot were nominally 0.25 mm diameter and were obtained from the same manufacturer. Two of the matched wire lots were given a special cleaning treatment by the manufacturer as part of a thermocouple development program under NASA Contract No. NAS 3-10950. The surface cleaning process, which includes ultrasonic degreasing, abrading, and electro-etching, was performed to more thoroughly remove surface contaminants introduced during the wire manufacturing processes. The third matched lot of wire was given no special cleaning by the manufacturer; however, all wires were cleaned with ether just before installation in the test facility.

Representative thermocouples from the matched lots were calibrated at 100 degree increments within the range 673 to 2073 K (400 °C to 1800 °C) by comparison with calibrated thermocouples and an optical pyrometer. These calibrations confirmed that the emf-temperature relationships of the "as received" thermocouples of each matched lot complied with the manufacturers standard calibration table (Ref. 4) to within ±1%. A check on the thermoelectric homogeneity of each spool of wire was also performed by determining the emf-temperature relationships between samples of "as received" wire taken from near the ends and middle of the spool. In every case the samples from the same spool were in agreement, thermoelectrically, to within 25 microvolts in the range 273 K to 2073 K (0 °C to 1800 °C).

A typical mass spectrographic analysis for the W-3% Re and W-25% Re wire lots is given in Table II. The W-3% Re wires typically had K, Al, Mo and Fe present in the range 300 to 50 ppmw, with other impurities totaling less than 100 ppmw. The W-25% Re wires typically had Mo, Si, Al, P and Fe present in the range 300 to 50 ppmw, K, Cr and Ni present at 20 to 30 ppmw, with other impurities totaling less than 100 ppmw.

TABLE II

TYPICAL MASS SPECTROGRAPHIC ANALYSIS OF WRe THERMOELEMENTS
(ppmw)

Element	W-3% Re Element	W-25% Re Element	Element	W-3% Re Element	W-25% Re Element
Li	0.4	1	Pd	< 0.2	< 0.2
Be	< 0.002	< 0.002	Ag	< 0.1	< 0.04
B	< 0.01	< 0.01	Cd	< 0.07	< 0.07
F	1	1	In	< 0.02	< 0.02
Na	10	10	Sn	< 0.07	0.1
Mg	1	1	Sb	< 0.05	1.5
Al	150	150	Te	< 0.1	< 0.1
Si	10	200	I	< 0.02	< 0.02
P	0.2	100	Cs	0.02	0.5
S	5	5	Ba	0.3	0.1
Cl	1	4	La	< 0.1	10
K	300	30	Ce	< 0.1	10
Ca	20	4	Pr	< 0.03	1
Sc	< 0.03	< 0.03	Nd	< 0.1	2
Ti (a)	< 15	< 15	Sm	< 0.1	< 0.1
V	1	1	Eu	< 0.06	< 0.06
Cr	3	20	Gd	< 0.1	< 0.1
Mn	0.1	2	Tb	< 0.03	< 0.03
Fe	50	50	Dy	< 0.1	< 0.1
Co	0.1	0.1	Ho	< 0.03	< 0.03
Ni	5	30	Er	< 0.1	< 0.1
Cu	0.6	2	Tm	< 0.03	< 0.03
Zn	< 0.4	< 0.4	Yb	< 0.1	< 0.1
Ga	< 0.04	< 0.2	Lu	< 0.03	< 0.03
Ge	< 0.1	< 0.1	Hf	< 0.1	< 0.1
As	< 0.01	1	Ta	2	2
Se	< 0.03	< 0.03	Os	< 0.07	< 0.07
Br	< 0.1	< 0.1	Ir	< 0.05	< 0.05
Rb	0.2	5	Pt (c)	< 3	< 3
Sr	0.2	0.05	Au (c)	$\underline{<}10$	$\underline{<}10$
Y	0.02	0.1	Hg (c)	< 5	< 5
Zr	< 3	< 3	Tl	< 0.4	< 1
Nb	(b)	(b)	Pb	< 1	< 1
Mo	120	200	Bi	< 1	< 1
Ru	< 0.06	< 0.06	Th	< 0.04	< 0.04
Rh	< 0.06	< 0.06	U	< 0.04	< 0.04

(a) Interference from Mo^{+N} ions

(b) Interference from W^{+N} ions

(c) Interference from matrix complexes - WC^{+}, WO^{+}, WOH^{+}, ReO^{+}, etc.

The Argon and Hydrogen gas used in the experiments were an ultra-high purity grade of commercially available compressed gas. The gases were certified by the supplier to contain less than 10 ppmv total impurities*, and within detectable limits, this was verified by NBS. The manufacturer's gas analyses for the gases as supplied in the gas cylinders are shown in Table III.

EXPERIMENTAL RESULTS

THERMOELECTRIC DATA

Most of the tests were performed with thermal exposure temperatures of 2400 K, and Table IV presents the changes in calibration that occurred after thermal exposure at 2400 K in the different environments for one of the matched wire lots. The ΔT (change in the calibration) at 2073 K is tabulated for the W-3% Re element, the W-25% Re element, and for a W-3% Re versus W-25% Re thermocouple in which both elements have been thermally exposed. The individual elements (and hence the composite thermocouples) which were tested in vacuum, Argon and Hydrogen all experience an initial change during the first 50 hours of the test. This initial change in calibration shall be designated as a shift.

For periods in excess of 50 hours at 2400 K, the thermoelements exposed in Argon and Hydrogen experience no further significant change in calibration beyond the initial shift, as shown in Table IV, where t_{max} is the maximum period of thermal exposure for a given environment. Typical curves showing the change in calibration of both thermoelements after being exposed in the gaseous environments are presented in Fig. 2, where the data is that for the Argon environment. Note that the change in thermal emf of each thermoelement is roughly linear with respect to temperature. The bands indicate the range of values obtained for thermoelements exposed for 50, 100, 250, 500, 750 and 1000 hours. No trend of the data within the bands with increasing exposure time was apparent. Any change in calibration which occurs after the initial shift shall be designated as a drift in calibration. Further, the initial period in which the calibration shift occurs shall be referred to as the thermal aging period. Thus, the thermoelements in Argon and Hydrogen experience no drift in calibration after thermal aging of the thermoelements is complete (and thus the shift has occurred).

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*On helium free basis in case of the Hydrogen.

TABLE III

ANALYSIS OF GASES CONTAINED IN GAS CYLINDERS

ARGON

Component	Composition in ppmv
O_2	< 5.0
N_2	< 5.0
CH_4	< 2.0
CO	< 1.0
CO_2	< 1.0
H_2	< 1.0
H_2O	< 4.0

HYDROGEN

O_2	< 1.0
N_2	< 5.0
CH_4	< 0.2
H_2O	< 6.0

TABLE IV

AVERAGE CHANGE IN CALIBRATION (ΔT IN KELVINS) AT 2073 K (1800 °C)
FOR THE THERMOELEMENTS AND THERMOCOUPLES FROM ONE MATCHED LOT
AFTER EXPOSURE AT 2400 K IN VACUUM, ARGON, AND HYDROGEN

ΔT IN KELVINS

ENVIRONMENT →	VACUUM (< 1 x 10^{-8} torr)			ARGON (1 atm)			HYDROGEN (1 atm)		
	W-3% Re Element	W-25% Re Element	Thermo-couple	W-3% Re Element	W-25% Re Element	Thermo-couple	W-3% Re Element	W-25% Re Element	Thermo-couple
SHIFT ΔT at 50 hours	+19	- 15	+ 4	+24	-7	+17	+16	-7	+ 9
OVERALL CHANGE ΔT at t_{max}	+21	-196**	-175**	+23	-6	+17	+19	-8	+11
DRIFT ΔT at t_{max} minus ΔT at 50 hours	+ 2	-181**	-179**	- 1	+1	0	+ 3	-1	+ 2
t^{*}_{max} →	500 hours			1000 hours			420 hours		

*t_{max} is the maximum exposure time in each environment.

**Large change due to Re vaporization in vacuum (see text page 15).

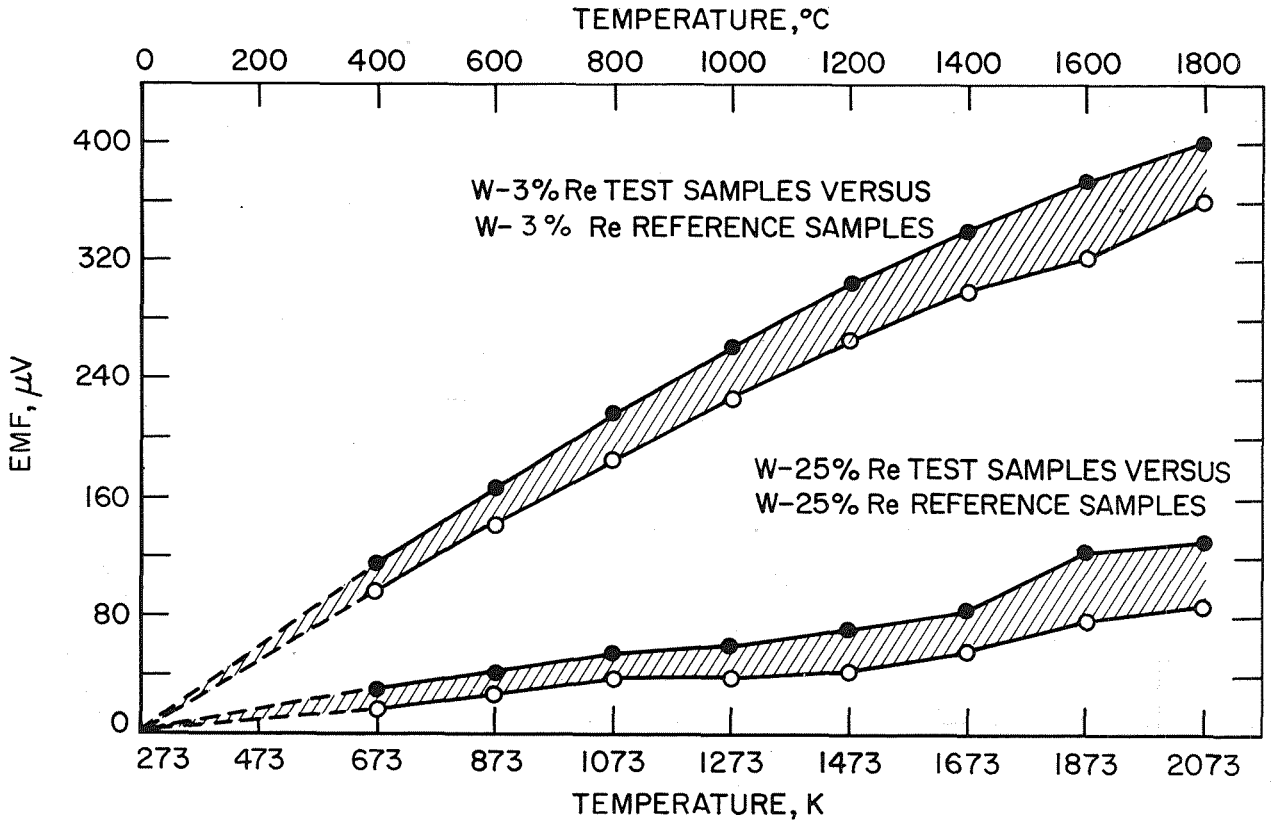

Fig. 2. Change in calibration (emf of thermally exposed thermoelement versus "as received" thermoelement from the same lot) for one lot of W-3% Re and W-25% Re thermoelements after thermal exposure at 2400 K in Argon for 50, 100, 250, 500, 750 or 1000 hours. All data points fell within cross-hatched areas, with no apparent trend with exposure time.

The long-term behavior of the thermocouple in the Argon and Hydrogen environments, illustrating the initial calibration shift followed by the absence of any significant calibration drift, is shown in Fig. 3. Again, this is for just one matched lot of material. The shift in calibration in Hydrogen appears to be slightly less (about 8 K) than that in Argon.

In a given environment, the shift in calibration will differ between wire lots. This is shown in Table V, which gives the changes in calibration at 2073 K for W-3% Re versus W-25% Re thermocouples from three different matched lots after thermal aging in Argon for 1 and 50 hours at 2400 K. Again, the changes in calibration of the individual elements is also given. The 1 hour test was included in order to verify that thermal aging would be complete in a time period much less than 50 hours at a thermal exposure temperature of 2400 K. Within experimental precision the shift in the calibration is complete after 1 hour at 2400 K.

In vacuum ($< 1 \times 10^{-8}$ torr), however, both the W-3% Re and the W-25% Re elements do experience a drift in calibration for exposure periods in excess of 50 hours at 2400 K. Although Table IV indicates that the W-3% Re thermoelement does not experience any appreciable drift in calibration at 2073 K after exposure for 500 hours in vacuum at 2400 K, in general, this is misleading. In Fig. 4a it is seen that a change in calibration does occur at lower calibration temperatures, and for the range 273 to 2073 K the drift in calibration is greatest at about 1000 K. For the W-25% Re thermoelement, the change in calibration within the range 673 to 2073 K is roughly proportional with calibration temperature, as shown in Fig. 4b.

The drift in calibration of the thermocouple experienced in vacuum is strongly dependent upon the temperature of exposure of the thermocouple as is illustrated in Fig. 5. The change in calibration at 2073 K for W-3% Re versus W-25% Re thermocouples is plotted as a function of time of exposure at wire temperatures of 2200 K, 2400 K, and 2600 K. Whereas the drift in calibration after exposure at 2200 K for 500 hours is negligible, at 2600 K a ΔT of more than 200 K occurs in 50 hours.

The drift in calibration of both the W-3% Re element and the W-25% Re element is a result of preferential loss of Re from the alloys by evaporation. Using electron microprobe analysis, the Re concentration was determined across the transverse cross-section of test wires at 10 or 20 micrometer intervals. The W-3% Re wires exhibited a decrease in Re concentration with increasing exposure time, and the Re concentration after 500 hours in vacuum at 2400 K

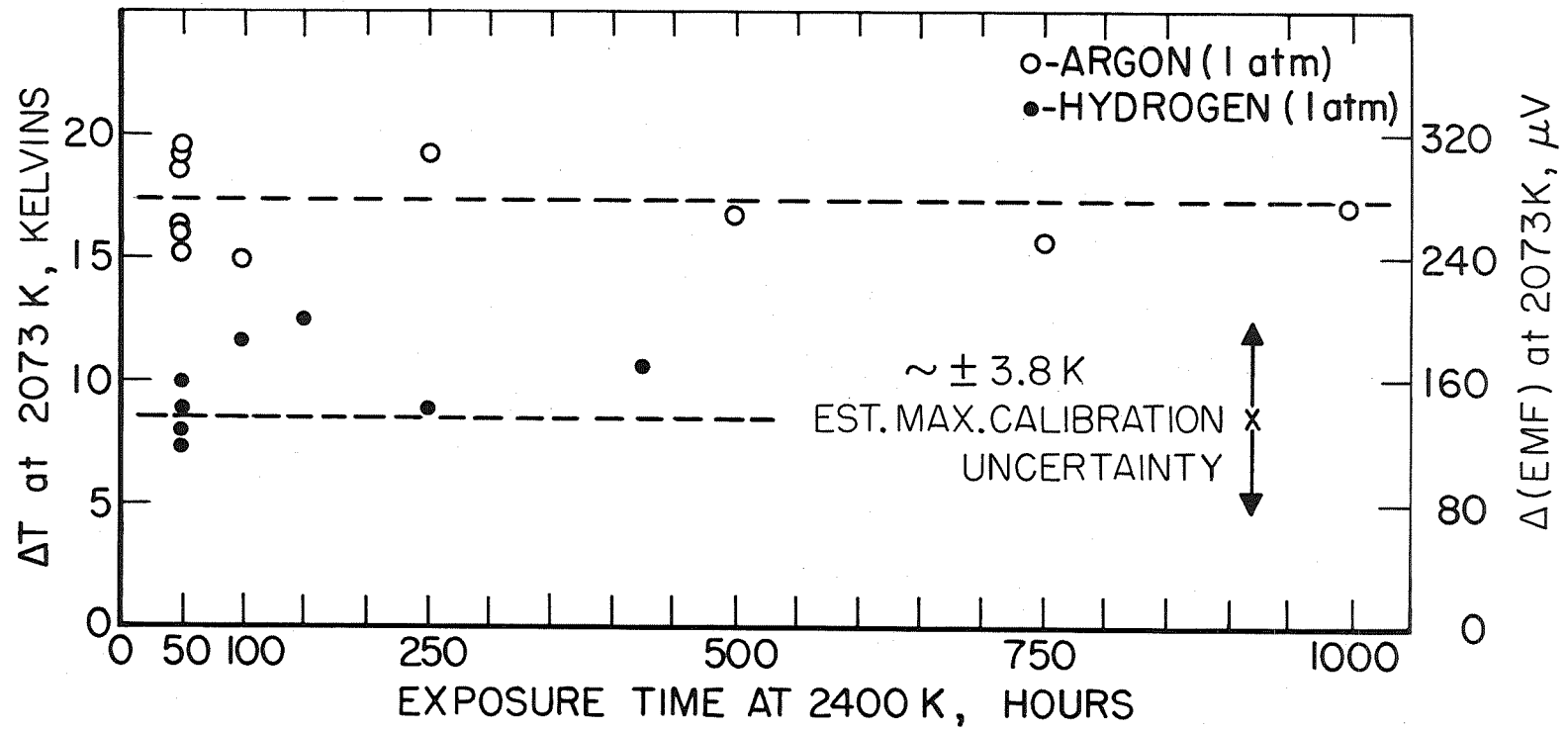

Fig. 3. Change in calibration (ΔT) at 2073 K of W-3% Re versus W-25% Re thermocouples from one matched lot as a function of exposure time at 2400 K in Argon or Hydrogen. Dashed curves indicate average change in calibration of thermocouples exposed for 50 hours (the shift).

TABLE V

AVERAGE CHANGE (SHIFT) IN CALIBRATION (ΔT IN KELVINS) AT 2073 K (1800 °C) OF THE THERMOELEMENTS AND THERMOCOUPLES FROM THREE DIFFERENT MATCHED LOTS AFTER EXPOSURE AT 2400 K IN ARGON (1 atm)

ΔT IN KELVINS

Exposure Time (Hours)	Specially Cleaned Lot			Specially Cleaned Lot			Uncleaned Lot		
	W-3% Re Element	W-25% Re Element	Thermo-couple	W-3% Re Element	W-25% Re Element	Thermo-couple	W-3% Re Element	W-25% Re Element	Thermo-couple
1	+6	-4	+2	+23	-5	+18	+9	-3	+6
50	+7	-4	+3	+24	-7	+17	+8	-2	+6

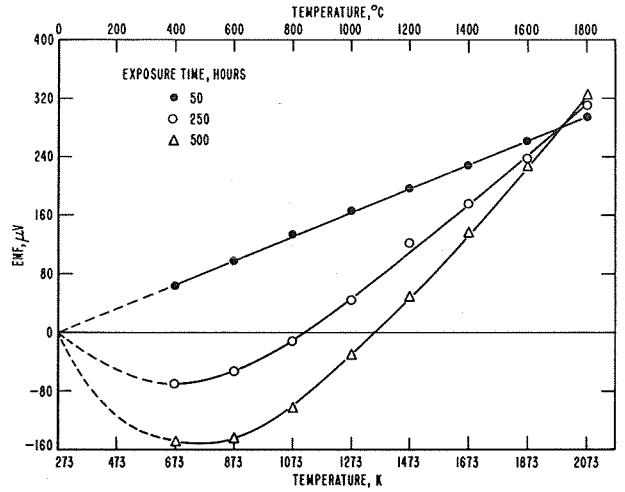

Fig. 4a. Change in calibration (emf of thermally exposed thermoelement versus "as received" thermoelement from the same lot) as a function of temperature for one lot of W-3% Re thermoelements after thermal exposure for 50, 250 and 500 hours at 2400 K in vacuum.

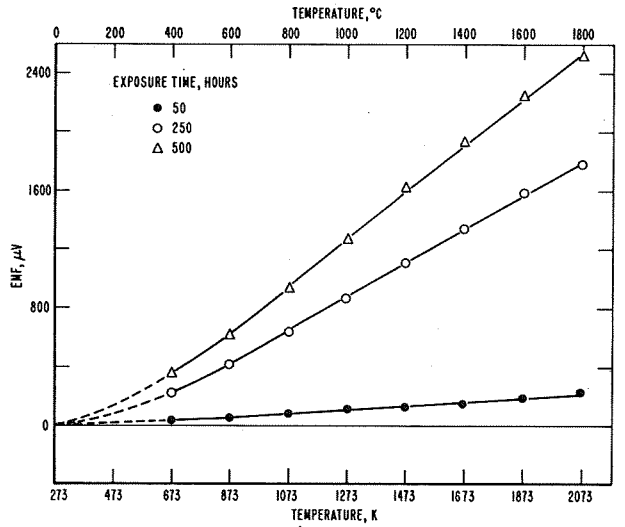

Fig. 4b. Change in calibration (emf of thermally exposed thermoelement versus "as received" thermoelement from the same lot) as a function of temperature for one lot of W-25% Re thermoelements after thermal exposure for 50, 250 and 500 hours at 2400 K in vacuum.

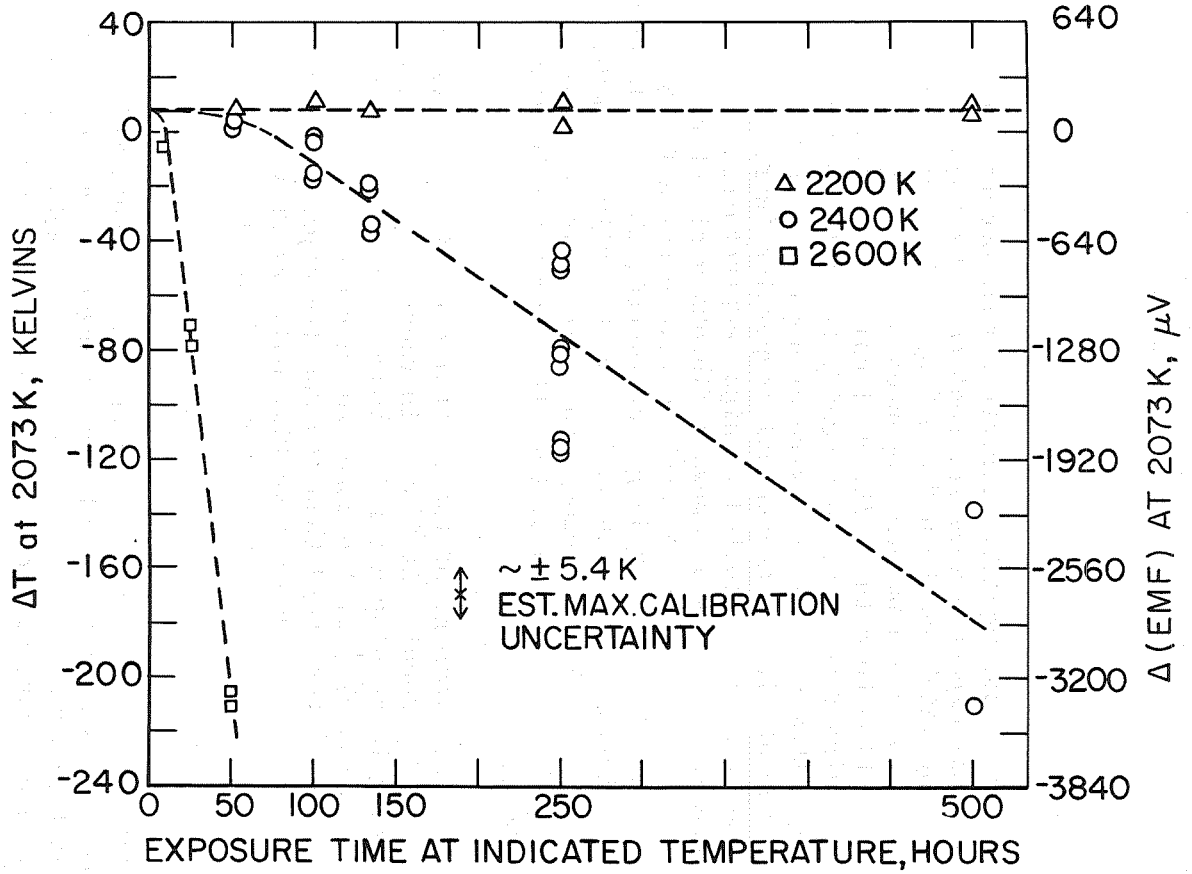

Fig. 5. Change in calibration (ΔT) at 2073 K of W-3% Re versus W-25% Re thermocouples from one matched lot as a function of exposure time in vacuum at 2200, 2400 and 2600 K. Dashed curves indicate trend of change in calibration (the drift).

had decreased from 3.0% to 2.5%. In addition, those wires subjected to the longer heating periods exhibited a slight decrease in Re concentration from the center of the wire to the surface. The W-25% Re wire heated 500 hours in vacuum at 2400 K exhibited a decrease from nominally 25% Re to 20.5% Re. At 500 hours exposure, the fractional loss of Re in both alloys is about 20%.

METALLURGICAL OBSERVATIONS

The changes in metallographic structure that occurred in the alloys after thermal exposure were interesting. The W-3% Re thermoelement is an alloy which has been "doped" by adding small amounts of potassium, silicon and aluminum compounds to the tungsten oxide prior to its reduction to metal. The residual doping elements which remain after the wire forming process modify the metallurgical structure changes that occur in the wire at high temperatures and result in improvements in the mechanical properties (Ref. 5). Before the "as received" wire is subjected to high temperatures, its microstructure is characterized by a somewhat fibrous grain structure, as illustrated in Fig. 6a. After the samples were exposed for 50 hours and longer at 2400 K in either Hydrogen or vacuum, they developed a grain structure similar to that shown in Fig. 6b, with a central core of very large interlocking elongated grains. The central core did not appreciably enlarge in diameter after the first 50 hours of exposure at 2400 K.

However, the development of the central core structure was found to be temperature dependent. At 2200 K in vacuum, the core structure was just beginning to develop after 250 hours of exposure; after 500 hours of exposure the size of the core had increased, but it was smaller in diameter (about 50%) than for samples exposed for periods of 50 hours and longer at 2400 K (see Fig. 6b). At 2600 K, the core structure was of about the same dimension as shown in Fig. 6b after 25 hours of exposure, and the core structure did not appreciably increase in size with 50 hours of exposure.

In contrast to the behavior in Hydrogen and vacuum, W-3% Re samples heated for as long as 1000 hours in Argon at 2400 K did not exhibit the central core structure. The change in metallographic structure that did occur, as shown in Fig. 6c, appeared as a slight amount of growth in the fine grain structure that took place during the first 50 hours of thermal exposure.

The change in metallographic structure of the W-25% Re thermoelement in all three environments was characterized by the growth of large equiaxed grains in the wire. The W-25% Re alloy is not "doped"

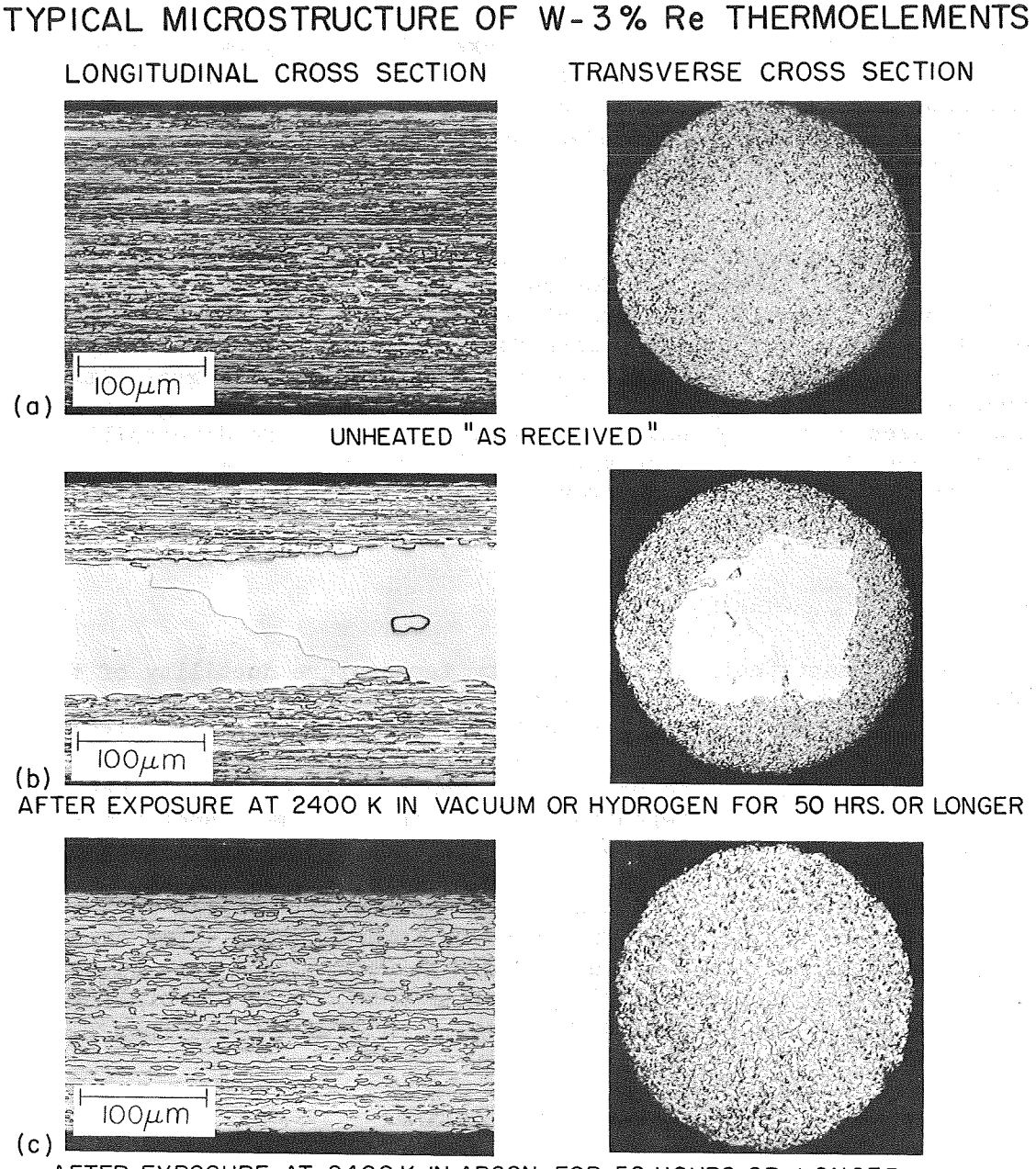

Fig. 6. Typical microstructure of W-3% Re thermoelements.

because the large Re concentration makes the grain growth control of these "doping" elements ineffective (Ref. 5). Attempts to test for long periods occasionally resulted in failure as the heated wire parted at a grain boundary under the influence of its own weight. This problem proved particularly troublesome in the Hydrogen environment.

Typical examples of the metallographic structure for the "as received" W-25% Re wire and for the W-25% Re wire after 1, 50 and 1000 hours of thermal exposure at 2400 K in Argon are given in Fig. 7. Samples exposed for one hour were fully crystallized and had experienced some grain growth. However, they had a decidedly finer grain structure than those samples exposed for the longer times. Further grain growth can be seen between 50 and 1000 hours. There were no discernible differences between the recrystallization behavior of W-25% Re samples exposed in the different environments as observed in longitudinal and transverse cross-sections.

DUCTILITY

While quantitative tests for room temperature ductility of the thermally exposed wires were not performed, an arbitrary measure of bend ductility was performed by coiling the wires on a 1 mm diameter mandrel. The W-3% Re wires exposed in Hydrogen and vacuum were extremely ductile, to the point where knots could be tied in the wire, and the wires could be repeatedly coiled on the 1 mm diameter mandrel and then straightened. The W-3% Re wires exposed in Argon were also ductile, but they were not as pliable as wires exposed in vacuum and Hydrogen. While they could usually be successfully coiled on the 1 mm diameter mandrel, they fractured on straightening. The exposed W-25% Re wires had large equiaxed grain structure, and were quite fragile, having only very limited room temperature bend ductility. It was not possible to coil them on the 1 mm diameter mandrel.

DISCUSSION OF RESULTS

RHENIUM LOSS IN VACUUM

The preferential loss of Re from the alloys when thermally exposed at high temperature in vacuum may be calculated, using classical kinetic theory and Raoult's law, as discussed by Dushman (Ref. 6). The calculation assumes that free evaporation occurs at the wire surface; that Raoult's law is valid; that changes in concentration are small; and that no concentration gradients exist in the sample. The calculation predicts that after 500 hours at

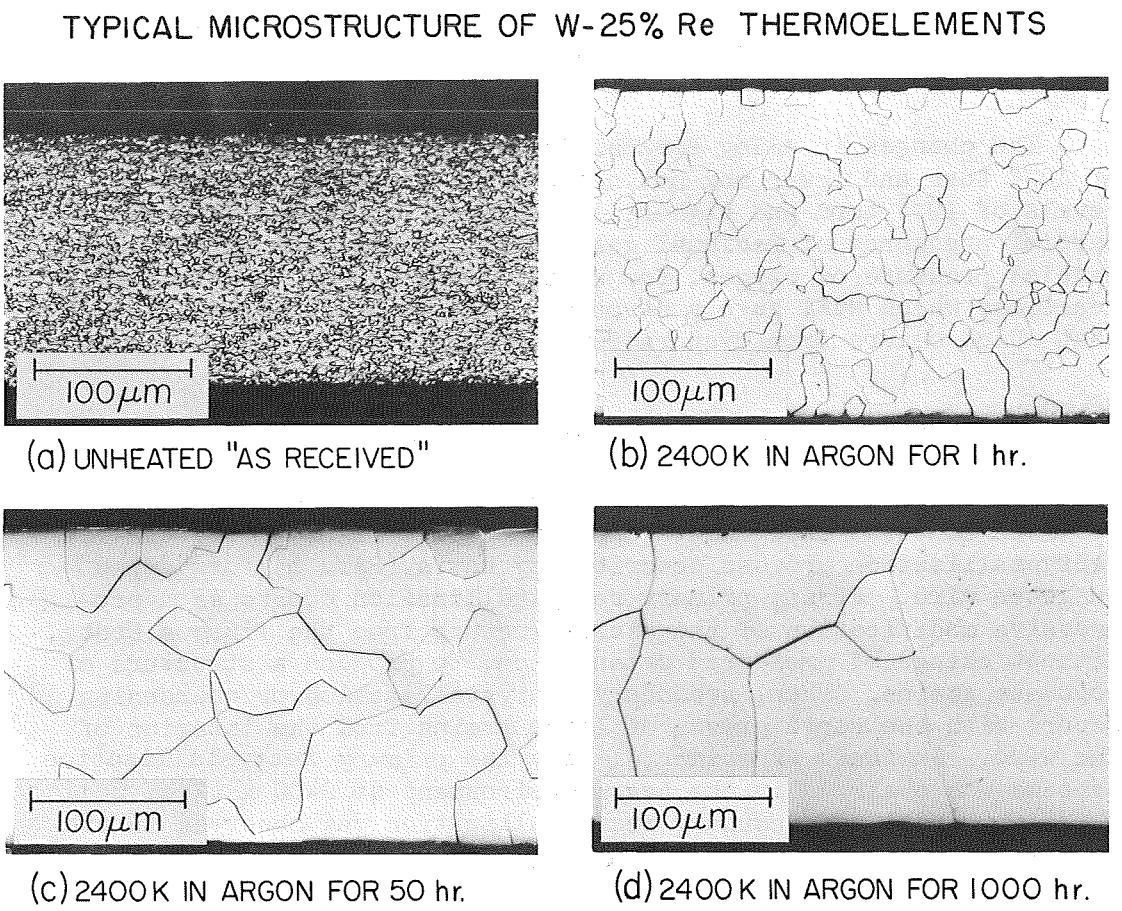

Fig. 7. Typical microstructure of W-25% Re thermoelements.

2400 K, an initial W-3.0% Re alloy would be W-2.7% Re, and an initial W-25% Re alloy would be W-23% Re. The values obtained experimentally were somewhat lower than this, but in view of the underlying assumptions and the large temperature dependence of the vapor pressure at these temperatures, the agreement of the calculated values with those found experimentally is considered most satisfactory.

The changes in alloy composition obtained at pressure levels of 10^{-8} torr and lower may not necessarily be valid at pressure levels of 10^{-5} torr and higher, depending upon the species and partial pressures of residual gases that are present. A high enough partial pressure of oxygen, for example, may result in chemical reactions which will have a stronger effect upon the alloys than that which occurs from evaporation (Ref. 7).

METALLURGICAL AND THERMOELECTRIC BEHAVIOR

The recrystallization behavior of the "doped" W-3% Re wire observed in these experiments appears to be similar to the two-stage recrystallization process described by Davis (Ref. 8) for "doped" tungsten wire. First, primary recrystallization occurs as a progressive modification of the microstructure from the fibrous state, without change of preferred orientation, to produce a structure of columnar grains. Then, secondary recrystallization or coarsening occurs with the rapid growth of large grains from the interior of the wire. In these experiments, only the primary recrystallization process was observed in the Argon environment at 2400 K (Fig. 6c), while the onset of secondary recrystallization was observed in the vacuum and Hydrogen environments (Fig. 6b).

The explanation for this phenomenon is not forthcoming. The role played by the additives (dopants) during the recrystallization processes is not well understood (Ref. 5 and 8). However, from the experiments a few positive statements can be made. First, it should be clear that the difference in the recrystallization behavior observed cannot be attributed to differences in the metallurgical, chemical, or thermal history of the particular starting materials used, since the tests were conducted with wire samples taken at random from the same lot (spool) of wire. It would seem unlikely that the difference in recrystallization behavior is a result of a difference in the temperature of exposure. The surface temperature of the wires during exposure was determined on a relative basis to within about ±25 K and was probably random from sample to sample within this range in each environment.

It may be that small differences in the transverse temperature gradients in the wire are responsible for the differences in recrystallization behavior. However, the heat losses at the wire surface in Argon are intermediate to that in vacuum and Hydrogen for the same exposure temperature, and therefore, one would not expect the Argon results to be anomalous to both the vacuum and Hydrogen results. In order to further pursue this last idea, a few tests were performed in a high purity Helium environment, where the heat losses at the wire surface are comparable with, but somewhat less than, that in Hydrogen. Samples exposed for 50, 100 and 250 hours in Helium at 2400 K did not exhibit secondary recrystallization and their microstructure was similar to samples exposed in Argon at 2400 K (Fig. 6c). Consequently, it appears that the recrystallization behavior of the "doped" W-3% Re material is dependent on the type of environment used. Of course, at thermal exposure temperatures above 2400 K, it is conceivable that secondary recrystallization can occur in the inert Argon and Helium environments as well; however, time and primary interests of the research did not permit further investigations.

At 2200 K in vacuum, the calibration shift for the W-3% Re wire is essentially completed within the initial 50 hour test period, yet the onset of secondary recrystallization (appearance of the central core) was only just beginning after 250 hours of exposure, and no abrupt change in the thermoelectric characteristics was evident with the onset of secondary recrystallization. Consequently, the shift appears to be essentially complete with primary recrystallization, and furthermore, no correlation appears to exist between the structural differences observed for W-3% Re wires exposed in the different environments and the small differences in the magnitude of the shift shown in Table IV. It may be that these differences in shift magnitude are related to small changes which might possibly occur in the physical or chemical behavior of the wire in the different environments during thermal aging.

The recrystallization behavior of the "undoped" W-25% Re wire is characterized by the formation of small equiaxed grains which then grow by consumption of each other. Since (see Table V) the shift is complete within the initial one hour exposure period in Argon, and thereafter the thermoelement does not drift, it is concluded that after recrystallization has occurred, subsequent grain growth (see Fig. 7) does not lead to any significant thermoelectric changes in the wire. Further, there were no discernible differences in recrystallization behavior of the W-25% Re wire in the different environments, nor did there appear to be any significant difference in the magnitude of the initial shift in emf of W-25% Re wires exposed in the different environments. (In vacuum at 2400 and 2600 K

the drift in emf of the W-25% Re element during the first 50 hours of thermal exposure is so large that it obscures the emf shift due to aging. However, in vacuum at 2200 K, where the drift is negligible, the shift in emf can be distinguished and within experimental precision it agrees with that found in Argon and Hydrogen at 2400 K for wire from the same lot).

The dependence of the shift upon the initial recrystallization has not been determined, since tests were not performed with exposure periods less than 5 hours at test temperatures of 2200 K and 2600 K, nor for less than 1 hour at 2400 K. It will be noted that other investigators have observed changes in the emf of W-Re thermocouple materials upon initial heating, and have recommended pre-annealing of these materials prior to use (Refs. 1 and 9). The existence of a shift in the calibration of the thermocouple wire is recognized by the thermocouple manufacturers, and part of their processing of the wire includes a heat treatment that tends to minimize the effect and yet maintain desirable mechanical properties.

CONCLUSIONS

1. W-3% Re versus W-25% Re bare wire thermocouples exposed at 2400 K in high purity Argon and Hydrogen experience no significant change in calibration beyond the initial shift in calibration for time periods up to 1000 hours.

2. Care must be taken with the use of these thermocouples in vacuum, since thermocouples exposed to temperatures of higher than 2200 K in high vacuum ($< 1 \times 10^{-8}$ torr) will drift in calibration as a result of the preferential loss of Re from the W-Re alloys. The drift in calibration is temperature and time dependent.

3. W-3% Re versus W-25% Re thermocouples undergo an initial positive shift in calibration of noticeable magnitude upon the first thermal exposure in vacuum or high purity Argon or Hydrogen. The thermal aging period required for the initial shift to be completed is probably much less than 50 hours at 2200 K and above; at 2400 K it is less than 1 hour.

4. The magnitude of the initial shift in calibration will vary for thermocouples from different lots, and will depend upon the environment in which the thermocouples are thermally aged. For the three matched lots tested, the magnitude of the initial shift in calibration was less than 20 K at 2073 K in all cases.

5. Large differences in the grain structure between the two alloys occur with thermal exposure. In the case of the W-3% Re alloy, grain growth was inhibited in the wire as a result of prior "doping", and the wire retained very ductile properties after thermal exposure. The W-25% Re alloy was not doped, and with thermal exposure exhibited large, equiaxial grain growth which resulted in brittle mechanical properties.

6. In the Hydrogen and vacuum environments, the W-3% Re alloy exhibited a core structure of very large interlocking grains (secondary recrystallization), whereas in the Argon environment, only broadening in the fine grain structure was evident (primary recrystallization). Most of the initial shift in calibration apparently occurs with primary recrystallization, but no relation between the magnitude of the initial shift and the onset of secondary recrystallization can be confidently assumed.

7. From examination of the grain growth in the W-25% Re element, it is evident that after recrystallization has occurred, the large subsequent grain growth does not lead to any significant thermoelectric changes in the wire.

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