CONF-8509153--2

ENGINEERING DATA BASES FOR REFRACTORY ALLOYS

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CONF-8509153--2

DE85 017714

ABSTRACT

Refractory alloys based on niobium, molybdenum, tantalum, and tungsten are required for the multi-100kW(e) space nuclear reactor power concepts that have been assessed in the SP-100 Program because of the extremely high temperatures involved. A review is presented of the technology efforts on the candidate refractory alloys in the areas of availability/fabricability, mechanical properties, irradiation effects, and compatibility.

Of the niobium-base alloys, only Nb-1Zr has a data base that is sufficiently comprehensive for the high level of confidence required in the reference-alloy selection process for the reactor concept to be tested in the Ground Engineering System (GES) Phase of the SP-100 Program. Based on relatively short-term tests, the alloy PWC-11 (Nb-1Zr-0.1C) appears to have significantly greater creep strength than Nb-1Zr; however, concerns as to whether this precipitation-hardened alloy will remain thermally stable during seven years of full-power reactor operation need to be resolved.

Additional information on the reference GES alloy will be needed for the detailed engineering design of a space power system and the fabrication of prototypical GES test components. Expedient development and demonstration of an adequate total manufacturing capability will be required if a high risk of significant schedule slippages and cost overruns is to be avoided.

INTRODUCTION

In the SP-100 Program concept design phase, Nb-1Zr and PWC-11 (Nb-1Zr-0.1C) were the refractory alloys designated by the design contractors for baseline and fallback designs, and T-111 (Ta-8W-2Hf), ASTAR-811C (Ta-8W-1Re-0.7Hf-0.025C), Mo-Re alloys, and W-Re alloys were indicated as candidates for the advanced designs. Reactor outlet temperatures ranged from 1100 to 1350 K for baseline and fallback designs and from 1000 to 1540 K for advanced designs.

In the concept selection process, the candidate alloys for these designs were carefully evaluated from the standpoint of their engineering data bases, with emphasis on availability/ fabricability of product forms and components, mechanical properties, radiation effects, and compatibility with coolants/working fluids. The purpose of this paper is to review and present the status of the SP-100 Program technology efforts on refractory alloys in these four areas.

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*Operated by Martin Marietta Energy Systems, Inc., for the U.S. Department of Energy under Contract No. DE-AC05-84OR21400.

AVAILABILITY/FABRICABILITY

The availability/fabricability of refractory-alloy product forms and components becomes an important consideration as the program advances to the engineering design and demonstration phase. Expedient development and demonstration of an adequate total manufacturing capability will be required if a high risk of significant schedule slippages and cost overruns is to be avoided.

A review of program activities in this area, along with some indications of existing U.S. industrial capabilities, is presented in this section. The product forms under consideration are characterized as plate, sheet and foil; pipe and tubing; and forgings. Examples of components are structural support grids, heat exchangers, vessels, pumps, and accumulators.

Product Forms. Activities on procurement and fabrication of refractory alloy product forms in the SP-100 Program have principally been in connection with supplying test specimens for the mechanical property and irradiation testing programs. A brief summary of these activities is as follows:

Alloy	Product Form(s) Fabricated
Nb-1Zr	Plate and sheet specimens from existing extruded sheet bar
PWC-11	Tubing, plate, and sheet from existing tubeshell extrusions
T-111	Tubing by cold-drawing existing tube stock
ASTAR-811C	Plate and sheet from sheet bar extrusions of procured arc-cast billets
Mo-13Re	Plate, sheet, and tubing from sheet bar and round bar extrusions of procured arc-cast billets
W-25Re	Plate and sheet from sheet bar extrusion of existing arc-cast ingot; small tubing from re-extrusion of 1-in. diam tubing

Only product forms of Nb-1Zr are considered to be readily available on a commercial scale. The number of current suppliers of refractory alloy product forms was found to be limited, there being three for Nb-1Zr and PWC-11 and only one each for T-111, ASTAR-811C, Mo-Re, and W-Re. Limiting factors in the fabrication of product forms are the chamber size of the annealing furnace and the degree of vacuum achievable. Brief descriptions of the vacuum annealing furnaces available in the U.S. commercial sector are given in Table 1. Pre-annealing qualification tests using protective getter foils will be required to utilize vacuum furnaces with the limited vacuum capability shown.

Components. Component fabrication has mainly to do with the joining together or welding of product forms, possibly with additional fabrication such as bending and spin-forming, and post-fabrication heat treatments as appropriate.

Table 1. Vacuum Annealing Furnaces Available in the U.S. Commercial Sector

Chamber Dimensions, in. Diameter	Chamber Dimensions, in. Length	Maximum Temperature (K)	Vacuum Capability (torr)
40	64	1800	1×10^{-4}
50	75	1650	5×10^{-5}
60	75	1800	1×10^{-5}
60	50	1850	5×10^{-5}
(a)	144	1800	1×10^{-5}
(b)	58	1900	1×10^{-5}

[a] Rectangular cross section 24 in. X 20 in.
[b] Rectangular cross section 48 in. X 28 in.

For the most part, large vacuum-pumped, inert-gas-purged gas-tungsten-arc (GTA) welding chambers are required for refractory-alloy component fabrication. The availability of such chambers in the U.S. commercial sector is summarized in Table 2. For SP-100 applications, refurbishment of these facilities will be required to meet the stringent impurity-level specifications of <2 ppm O_2 and <10 ppm H_2O, and custom-built chambers probably will be required for final assembly of large ground engineering system or flight system components. GTA field welding equipment, such as that developed in the past for fabricating refractory alloy components, does not currently exist, and electron beam welding capability is limited to specific geometries.

Table 2. Large Vacuum-Pumped, Inert-Gas-Purged GTA Welding Chambers Available in the U.S. Commercial Sector

Number of Units	Chamber Dimensions, in. Diameter	Chamber Dimensions, in. Length
1	72	126
1	48	60
2[a]	36	78
	60	76
1	48	60
10	60	24

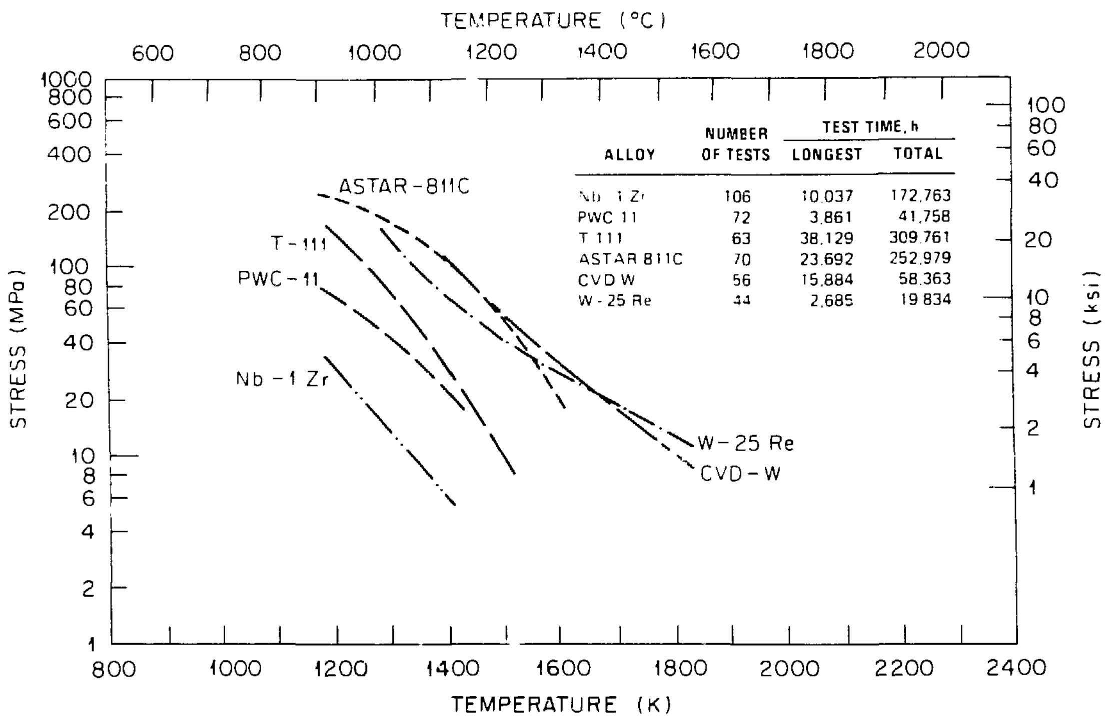
[a] Attached chambers.

Optimized procedures for welding refractory alloy components are available only for Nb-1Zr and T-111, although no major problems are anticipated for PWC-11 and ASTAR-811C. Welding experience is very limited for Mo-Re and W-Re alloys. Except for Nb-1Zr and T-111, procedures for postweld heat treatments must be developed and/or optimized. In all cases, training and certification of personnel will be required.

MECHANICAL PROPERTIES

Considerable progress has been made during the past year on understanding the creep behavior of candidate refractory alloys. This has been accomplished by more thoroughly analyzing previously reported data as well as performing additional creep tests on selected alloys.

The current situation in terms of the estimated stress to produce 1% creep strain in 7 years based on extrapolations using the Larson-Miller parameter approach (with arbitrary constant of 15) is shown in Fig. 1. A measure of the relative reliabilities of these extrapolations is afforded by the information shown in the upper right hand portion of the figure on the number of tests performed, the longest single test, and the total test time for each alloy.



ALLOY	NUMBER OF TESTS	TEST TIME, h	
		LONGEST	TOTAL
Nb 1 Zr	106	10.037	172,763
PWC 11	72	3.861	41,758
T 111	63	38.129	309.761
ASTAR 811C	70	23.692	252.979
CVD W	56	15,884	58.363
W-25 Re	44	2.685	19 834

Fig. 1. Stress to produce 1% creep strain in 7 years for candidate SP-100 refractory alloys estimated from extrapolations using the Larson-Miller parameter with an arbitrary constant of 15.

The alloys represented in this plot are only those for which (a) reasonably comprehensive data bases exist; (b) the test environments were known to be of high quality, i.e., either lithium or vacuum-test-chamber pressures of 10^{-7} torr or less; and (c) the proper processing and heat treating procedures were used. For example, some preliminary results on W-5Re and Mo-11/14Re, for which the longest single test times were 89 and 263 h and the total test times were 96 and 579 h, respectively, are not shown.

Based on data from tests of several thousands of hours duration, the creep strength of properly heat-treated PWC-11 appears to be significantly greater than that of Nb-1Zr. There is evidence, however, that this precipitation-hardened alloy overages owing to coarsening of the Nb-Zr carbide precipitates at proposed system operating temperatures. Tests to tens of thousands of hours of properly processed and heat-treated material are needed to determine whether PWC-11 will retain its apparent creep-strength advantage over Nb-1Zr for the goal SP-100 mission life of seven years at full power.

Tests on PWC-11 at the NASA-Lewis Research Center (NASA-LeRC) by R. H. Titran and T. J. Moore showed that specimens containing electron-beam welds exhibited significantly less creep strength than unwelded specimens. Creep testing of PWC-11 at NASA-LeRC is continuing.

Creep tests are in progress at the Oak Ridge National Laboratory (ORNL) on three separate heats of Nb-1Zr, with specimens containing both longitudinal and transverse electron-beam welds, at 1250-1550 K and on CVD-W at 1700 K. The test conditions were selected to produce 1% creep in several thousand hours.

Some interest has been shown recently in considering the possibility of employing a currently commercial, readily fabricable niobium-base alloy known as C-103 (Nb-10Hf-1.0Ti-0.7Zr-0.5Ta) as a stronger substitute for Nb-1Zr. However, careful analysis of extensive creep data on this alloy reported by Titran and Klopp[1] and summarized by Conway[2] indicates that C-103 holds no advantage over Nb-1Zr from the standpoint of creep strength for SP-100 applications.

It is generally agreed that only the creep data bases for Nb-1Zr, T-111, and ASTAR-811C are adequate for engineering design purposes. Additional data are needed, however, on weldments and the effects of heat-to-heat variations. In general, the behavior of the candidate alloys under conditions of alternating stresses, creep fatigue, and multiaxial loadings also must be assessed. In addition, fracture toughness and crack-growth rate studies are needed to assess the potential for brittle failure of structural components under anticipated launch and orbital-boost conditions.

IRRADIATION EFFECTS

The current effort on assessing the effects of irradiation on the properties of candidate refractory alloys in the SP-100 Program is described in the paper by Cox et al. elsewhere in the proceedings of this meeting. Two aspects not covered in that paper are (1) the results of tensile tests performed at ORNL on Nb-1Zr and PWC-11 specimens irradiated in the HEDL/EBR-II program and (2) the results of tensile tests and swelling measurements on specimens of unalloyed tantalum and three tantalum alloys from EBR-II experiments performed in an ORNL program about a decade ago.

Tensile Properties of Irradiated Nb-1Zr and PWC-11. Irradiation test conditions for the Nb-1Zr and PWC-11 specimens in the HEDL/EBR-II program were as follows: environment, lithium; peak fluence, 1.9×10^{26} neutrons/m^2 (E > 0.1 MeV); estimated temperature, 1600 K. The tensile tests were performed under vacuum (about 10^{-6} torr) at 1550 K by M. L. Grossbeck.

The results of the tests are presented in Table 3. Note that both alloys exhibited significant decreases in strength and increases in ductility, possibly as the result of the transfer of oxygen to the surrounding lithium during the irradiation tests. For PWC-11, these effects may also be associated with overaging (the precipitation-hardening heat treatment temperature was 1480 K, i.e., somewhat less than the exposure and test temperatures).

Table 3. Summary of Results of Tensile Tests at 1550 K on Nb-1Zr and and PWC-11 Specimens Irradiated in EBR-II at 1600 K to Peak Fluence of 1.9×10^{26} neutrons/m^2 (E > 0.1 MeV)

	Strength, MPa				Elongation, %			
	Yield		Tensile		Uniform		Total	
Alloy	Before	After	Before	After	Before	After	Before	After
Nb-1Zr	54	37	67	57	10	32	56	67
PWC-11	60	47	74	67	11	20	37	54

Previously Irradiated Tantalum Alloys. Specimens of unalloyed tantalum and the tantalum alloys Ta-10W, T-111, and ASTAR-811C were irradiated in the EBR-II at temperatures of 660–920 K to a fluence of 1.7×10^{26} neutrons/m^2 (E > 0.1 MeV) about a decade ago as part of the breeder reactor control materials development program. Because of lack of interest in these materials for that application, the irradiated specimens were not examined but were placed in storage.

The swelling characteristics and tensile properties of these specimens were determined recently by Grossbeck and Wiffen.[3] The tensile test temperatures were the same as the irradiation temperatures, the pressures ranged from 5×10^{-8} to 1×10^{-6} torr, and the strain rate was 3×10^{-4}/s. The results are summarized below.

- Unalloyed tantalum swelled by 0.36%, but swelling in the alloys was negligible.
- T-111 showed as much as a three-fold increase in yield strength but, along with Ta-10W, exhibited gross plastic instability, with less than 0.2% uniform elongation.
- For ASTAR 811C, the yield strength increased by as much as a factor of 2, and no plastic instability was observed.
- Considerable hardening and reduction in ductility, but no plastic instability, was observed in the irradiated specimens of unalloyed tantalum.
- T-111 doped with 400 wt ppm (4500 at. ppm) oxygen and irradiated at 753 K was severely embrittled (no plastic deformation in the tensile test); scanning electron microscopy of fracture surfaces revealed severe cleavage and intergranular fracture.

The absence of plastic instability in ASTAR-811C is considered to be encouraging not only for this alloy but for other alloys containing fine carbide precipitates. These precipitates may prevent channeling (the phenomenon whereby dislocations sweep through a narrow band free of dislocation loops) which leads to plastic instability.

COMPATIBILITY

In general, the refractory alloys have been shown to be very compatible with alkali metals.[4] During the past year, additional studies in alkali metals were performed, and some tests in helium also were conducted.

Alkali Metals. Exposures of unalloyed molybdenum, Mo-Re alloys, and Nb-1Zr to lithium and sodium environments in the form of heat-pipe life tests have been conducted at the Los Alamos National Laboratory. The results of these tests are reported by Merigan in another paper in the proceedings of this meeting.

Helium. T. J. Moore and C. M. Scheurmann of NASA-LeRC performed studies on the temperature-gradient mass transport of carbon, nitrogen, and oxygen in Nb-1Zr systems circulating helium under simulated Brayton cycle and Stirling engine conditions. The extent of mass transport was determined from Auger profiles and by chemical analyses of Nb-1Zr specimens.

In the simulated Brayton cycle experiments, Nb-1Zr coupons were exposed to helium in a 770 K/1300 K temperature gradient in a molybdenum capsule for 748 h. Under these conditions, no interstitial mass transport was detected. In the experiment simulating Stirling engine conditions, coupons of Nb-1Zr, Al_2O_3, SmCo, and Hyperco steel were exposed in a molybdenum capsule to helium in a 750 K/1300 K temperature gradient for 508 h. Some evidence of the transport of carbon and oxygen to the Nb-1Zr coupons in the hot zone was found.

Based on the results of these initial tests, it was recommended that additional experiments be performed for longer times and under conditions that more closely simulate Brayton and Stirling systems.

SUMMARY AND CONCLUSIONS

The technology efforts on candidate refractory alloys for SP-100 applications have been reviewed with emphasis on the areas of availability/fabricability, mechanical properties, irradiation effects, and compatibility.

In the area of availability/fabricability, expedient development and demonstration of an adequate total manufacturing capability will be required as the program advances to the engineering design and demonstration phase if a high risk of significant schedule slippages and cost overruns is to be avoided.

The creep data bases for Nb-1Zr, T-111, and ASTAR-811C appear to be adequate for engineering design purposes, although additional data on weldments and the effects of heat-to-heat variations are needed. In general, additional data are needed on fatigue, creep-fatigue interactions, effects of multiaxial stress states, fracture toughness, crack-growth rates, and irradiation effects. Based on relatively short-term tests of thousands of hours, PWC-11 appears to have significantly greater creep strength than Nb-1Zr; however, concerns as to whether this precipitation-hardened alloy will remain thermally stable during seven years of full-power reactor operation need to be resolved.

All factors considered, only the Nb-1Zr data base appears to be sufficiently comprehensive for the high level of confidence required in the reference niobium-base-alloy selection process for the reactor concept to be tested in the Ground Engineering System (GES) Phase. However, additional information on this alloy is needed for the detailed engineering design of a space power system and the fabrication of prototypical GES test components. To this end, technology programs are planned for the qualification of manufacturing methods for product forms and components, the development of high-temperature design methods, the enhancement of mechanical properties information, and further evaluation of irradiation effects.

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