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**SHORT TERM CREEP RUPTURE PREDICTIONS
FOR TANTALUM ALLOY T-111***

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Abstract

A knowledge of the short term creep rupture behavior of Tantalum alloy T-111 is necessary to predict device integrity in the heat source section of Radioisotope Thermoelectric Generators (RTG's) at the end of service life, in the event of a fuel fire. High pressures exist in RTG's near the end of service life, these are caused by gas generation resulting from radioactive decay of the nuclear fuel. The internal pressure exerts a significant hoop stress on the T-111 alloy structural containment member. This paper analyses the short term creep behavior (rupture times up to $\sim 2 \times 10^3$ hrs.) of cold worked (CW) T-111 alloy, using the existing data of Stephenson (1967). Correlations for the time to rupture, time to 1% strain and minimum creep rate have been obtained from this data using multivariable linear regression analysis. These results are compared to other short term rupture data for T-111 alloy. Finally, at the stress/temperature levels relevant to the RTG fuel fire scenario near the end of service life, the rupture time correlation for T-111 alloy predicts a rupture time of approximately 100 hrs.

INTRODUCTION

Tantalum alloy T-111 (nominal composition: Ta-8wt %W-2Hf) is currently used as the structural containment member for nuclear fuel in the heat source section of Radioisotope Thermoelectric Generators (RTG's). These components have been used during the past 20 years in various applications where a reliable, long life power supply is required. While the normal operating temperature of the RTG heat source (~ 723 K) is easily within the range of application of nickel base alloys, the requirements for containment of nuclear fuel in the event of a fuel fire leads to the use of refractory alloys such as T-111 which have good mechanical properties at temperatures > 1300 K. For the RTG's of current interest, a fuel fire would require the structural containment member to withstand a maximum temperature of 1333 K for up to 2 hrs. During the 20 year service life of a RTG, there is a buildup of internal gas pressure in the heat source due to radioactive decay of the nuclear fuel. Thus, it is most important to verify that the pressure buildup at the end of service life (36.5 MPa maximum) would not rupture of the containment vessel. The extreme conditions involved during a fuel fire scenario indicate that one must be able to show that the creep rupture properties of the T-111 alloy containment member are sufficient to assure containment.

This paper summarizes the results of a study (Stephens, 1989) which developed short term creep rupture and minimum strain rate relations for T-111 alloy. Since the T-111 used for the containment vessel is in the cold worked (CW) plus stress relief annealed condition, we have focused primarily on creep studies of T-111 alloy performed using specimens in the CW, as opposed to recrystallized, condition.

SURVEY OF UNIAXIAL CREEP-RUPTURE DATA FOR T-111 ALLOY

T-111 alloy was developed for long term creep resistance at elevated temperatures, i.e.,

conditions appropriate for strain-limited applications in space nuclear power reactors. As such, there is a relatively large long term creep data base available for this alloy, performed in ion pumped creep frames at pressures of $\sim 1.3 \times 10^{-6}$ Pa (10^{-8} Torr). This data is tabulated in Conway and Flagella (1971) as well as in Sheffler, et. al. (1969, 1973). The majority of the long term data is for relatively low stresses, long times (10^3 - 10^4 hrs.) and the tests were terminated at fairly low creep strains (2-4% strain). This is understandable insofar as most of the space reactor applications are strain-limited as opposed to creep-rupture limited. For example, Horak (1987) indicates that for the SP-100 program, the major creep design parameter is the stress at a given temperature to produce 1% creep strain over the mission life of 7 years.

The most complete study of short term uniaxial creep-rupture behavior for T-111 alloy is that of Stephenson (1967). Creep-rupture data for T-111 alloy in the cold worked condition were obtained at temperatures of 1477, 1700, and 1922 K. The chemical composition of the heat of T-111 used for the tests is shown in Table 1. The tests were performed in diffusion pumped vacuum creep machines with pressures $< 2.6 \times 10^{-5}$ Pa (2×10^{-7} Torr). The rupture time (t_r) data obtained ranged from 0.5 up to 2000 hrs. In addition to t_r , data for time to 1% strain ($t_{1\%}$) and minimum strain rate ($\dot{\epsilon}_{\min}$) were also plotted for the various tests. Figures 1 and 2 show the data for t_r and $\dot{\epsilon}_{\min}$ from Stephenson (1967) which were used for analysis in this study.

There is also a limited amount of additional short term creep-rupture data available on T-111 alloy. Stoner and Buckman (1970) performed a number of tests at 1588 K on T-111 alloy tubing using ion pumped vacuum creep machines at pressures of $\sim 1.3 \times 10^{-6}$ Pa (10^{-8} Torr). Three different conditions were examined, including as-drawn seamless tubing, welded plus drawn tubing and as-longitudinally welded material; there did not appear to be any significant difference in rupture time for these various conditions. The composition of the T-111 heat used for these tests is also shown in Table 1. There is also a limited amount of rupture time data for recrystallized T-111 alloy in Schmidt and Ogden (1963); these data were apparently from programs conducted by Westinghouse for the U.S. Navy as part of the original alloy development work on T-111. The rupture time data from these two studies are plotted in Figure 3.

Table 1 compares the chemical compositions of two recent lots of T-111 alloy purchased for RTG fabrication to the T-111 used in the study of Stephenson (1967) and Stoner and Buckman (1970). It is apparent from inspection of Table 1 that the level of both major alloying elements and trace elements of lots 11 and 12 are indeed comparable to those heats used for creep testing in both of the above studies.

RESULTS

Using multivariable linear regression analysis procedures (Anderson, 1982), the uniaxial time to rupture data for CW T-111 alloy from Stephenson (1967) were fit to the following power law correlation incorporating stress and temperature:

$$t_r \text{ (hrs.)} = 2.02 \times 10^{-2} \sigma^{-5.45} \exp(4.475E5/RT) \quad (1)$$

In Equation (1), t_r is the rupture time (hrs.), σ is the applied uniaxial stress in MPa, R is the gas constant (8.313 Joules/mole K), and T is the temperature in Kelvin. The lines drawn in Figure 1 show the predicted values of t_r for the three temperatures which Stephenson investigated, i.e., 1477, 1700 and 1922 K. Inspection of Figure 1 indicates that Equation (1) provides an excellent fit to the experimental data. This is consistent with the correlation coefficient (r^2 parameter) for Equation(1), which was 0.973. Using an equation with the same functional form, the data for time to 1% strain ($t_{1\%}$) for CW T-111 alloy were analysed, with the following results:

$$t_{1\%} \text{ (hrs.)} = 1.76 \times 10^{-2} \sigma^{-3.65} \exp(2.928E5/RT) \quad (2)$$

TABLE 1. Chemical compositions of two heats of T-111 alloy used for RTG fabrication, along with analysis of the heats studied by Stoner and Buckman (1970) in NASA CR-72869 and Stephenson (1967) in ORNL TM-1994. A dash mark indicates a range of compositions were measured for various samples of the same heat, while a slash mark indicates the results of two independent measurements.

Element	Lot 11	Lot 12	NASA CR-72869	ORNL TM-1994
W (wt.%)	7.2-8.1*	7.96/8.05**	8.1-8.2	7.4/7.5
Hf (wt.%)	1.8-1.9	1.99/2.07	1.86-2.10	2.1/1.9
Nb (wppm)	<100	70/117	<100	n.a.
Mo (wppm)	<50	<100	n.a.	n.a.
Ni (wppm)	<50	<50	<10	n.a.
O (wppm)	37-48	34/54	40-44	57/53
C (wppm)	14-32	19/31	10-50	49/70
N (wppm)	10-13	14/15	10-40	23/29
H (wppm)	7-8	3/8	1-7	n.a./4
Zr (wppm)	300	370/384	200-1000	n.a.
Ta	bal	bal	bal	bal

FIGURE 1. Time to rupture as a function of applied stress for cold worked T-111 alloy, data of Stephenson (1967). Solid lines indicate the predicted rupture times for 1477, 1700 and 1922 K obtained using Equation (1).

The correlation coefficient obtained for Equation (2) was 0.906.

A comparison of the predicted rupture times at 1588 K and the data from both Schmidt and Ogden (1963) and the work of Stoner and Buckman (1970) is shown in Figure 3. It is apparent that fairly good agreement is obtained between these data and the rupture time correlation presented in Equation (1).

The minimum strain rate data for CW T-111 alloy were also analysed using multivariable linear regression analysis techniques. The following correlation was obtained:

$$\dot{\epsilon}_{\min} (\text{s}^{-1}) = 2.18 \times 10^{-3} \sigma^{4.51} \exp(3.872\text{E}5/\text{RT}) \quad (3)$$

As in the case of the correlation for t_r , a good correlation coefficient was obtained for Equation (3), with $r^2 = 0.938$. The agreement between the T-111 data and Equation (3) is shown in Figure 2. It is interesting to note that the apparent activation energy for creep obtained in Equation (3), 387.24 kJ/mole (92.55 kcal/mole), is very close to the apparent activation energy used by Sheffler, et. al. (1969), 376.56 kJ/mole (90.0 kcal/mole), to plot minimum strain rate data for recrystallized T-111 alloy. Both values of apparent activation energy are less than the activation energy for self diffusion of Ta (413 kJ/mole = 98.7 kcal/mole) recommended by Frost and Ashby (1982). If creep tests were conducted at higher temperatures on T-111 alloy, one would expect the activation energy for creep to either equal or slightly exceed the activation energy for self-diffusion in Ta.

FIGURE 2. Minimum strain rate as a function of applied stress for cold worked T-111 alloy, data of Stephenson (1967). Solid lines indicate the predicted minimum strain rates for 1477, 1700 and 1922 K obtained using Equation (3).

One can obtain a predictive relation for the creep strain to fracture to be expected for CW T-111 alloy by means of evaluating the product of $\dot{\epsilon}_{\min}$ and t_r to obtain the Monkman-Grant parameter. This parameter, with units of strain, provides a rough estimate of the uniaxial strain to fracture for a given alloy. By combining Equations (1) and (3), it is possible to obtain an expression for the Monkman-Grant parameter, C_{mg} , for CW T-111 alloy:

$$C_{mg} = \dot{\epsilon}_{\min} * t_r = 1.59 \times 10^{-1} \sigma^{-0.94} \exp(6.026E4/RT) \quad (4)$$

Thus, there is a slight temperature/stress dependence to C_{mg} for CW T-111 alloy - as is usually encountered in engineering alloys. For the data obtained at the highest stress and lowest temperature (241.3 MPa, 1477 K) examined by Stephenson, Equation (4) predicts a value of 0.124. This value of C_{mg} is the minimum expected over the range of temperature and stress examined by Stephenson and suggests that T-111 alloy is not susceptible to intergranular fracture under these conditions.

FIGURE 3. Time to rupture as a function of applied stress for T-111 alloy at 1588 K, recrystallized data (triangles) from Schmidt and Ogden (1963), while the data from Stoner and Buckman (1970) shown with plus signs, are for T-111 tubing in various conditions (see text). Solid lines indicate the predicted rupture times for 1333 and 1588 K obtained using Equation (1).

RUPTURE TIME PREDICTIONS: FUEL FIRE SCENARIO

As indicated above, the RTG assembly would be expected to see a maximum temperature of 1333 K for 2 hours as a result of a fuel fire. Given a maximum internal pressure of 36.5 MPa for the RTG's of interest, hoop stresses as high as 346.8 MPa (50.3 ksi) could be expected on the T-111 containment member at the end of service life. A plot of the predicted uniaxial rupture time as a function of stress for 1333 K using the correlation shown in Equation (1) is shown in Figure 3. A rupture time of 100.1 hrs. is predicted for 1333 K/346.8 MPa, while a minimum

strain rate of $4.14 \times 10^{-7} \text{ s}^{-1}$ is predicted using Equation (3). The value of C_{mg} for these same conditions is thus 0.149, which suggests a significant amount of creep strain would be observed prior to fracture. Finally, the time to 1% strain is predicted by Equation (2) to be 2.82 hrs. Thus, if a fuel fire occurred near the end of service life, the T-111 heat source containment member would be expected to undergo some creep deformation, but only a very small portion of the total strain to fracture.

A "time independent" prediction regarding the strength of the T-111 containment could also be made with the elevated temperature tensile data for recrystallized T-111 alloy presented by Sheffler, et. al. (1969). Their data indicate that at 1333 K recrystallized T-111 would have a 0.2% offset yield strength of approximately 234.4 MPa, along with an ultimate tensile strength of 441.3 MPa and 20% elongation. Thus, one would also conclude from this tensile data that the T-111 heat source containment member will plastically deform, but the stresses involved would be significantly less than the UTS at 1333 K.

There is one important aspect of the above results that should be emphasized: we have used uniaxial creep data to predict stress rupture in a cylindrical geometry, rather than the uniaxial conditions under which all of the creep rupture data were generated. While it is likely that the hoop strain to fracture will be somewhat less than the uniaxial strain to fracture, there appears to be sufficient creep ductility in T-111 alloy that this is not a problem. The cylindrical hoop stress conditions also indicate that the minimum strain rate will be lower than predicted by the uniaxial correlation. By consulting Odqvist (1974), it can be shown that for the value of stress exponent obtained in Equation (3), i.e., 4.51, the creep rate caused by a hoop stress would be expected to be a factor of 2.2 less than the uniaxial creep rate at the same (uniaxial) stress level. One would thus expect the use of uniaxial rupture time data to be conservative by a similar amount when considering the cylindrical (hoop stress) loading conditions.

CONCLUSIONS

This study has demonstrated that a reasonable short term minimum creep rate, time to 1% strain and rupture time correlations can be obtained for CW T-111 alloy using the data of Stephenson (1967). Rupture time data obtained at 1588 K for other heats of T-111 which were examined by other laboratories are in good agreement with the rupture time correlation presented in Equation (2). We have used the various correlations to verify that at the stress levels expected near the end of service in an RTG heat source, the T-111 alloy has sufficient creep rupture strength to withstand the fuel fire scenario of 1333 K exposure for 2 hours. In particular, these correlations predict that at the uniaxial stress level of 346.8 MPa, that the time to 1% strain is predicted to be 2.82 hours, while the time to rupture is 100.1 hrs. The fact the stresses are in fact cylindrical hoop stresses tends to reduce the creep rate of the alloy - and thereby one would expect the 100.1 hr. rupture time prediction to be somewhat conservative.

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