Creep to Leakage of Pt-20Rh:
The AI Report and Other Comments

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SUMMARY

The data presented in the recently issued AI report on Pt-20Rh creep are discussed. The data consist of six values and differ significantly from the four data points that have been attributed to the AI report. If all six points are used, the scatter is so large that application of the usual normal distribution model is questionable and leads to the prediction of low reliabilities.

A mechanism of capsule failure by leaking is presented that includes the random occurrence of leak paths and predicts a high degree of scatter in results. If this mechanism is correct, results should be analyzed by the binomial model rather than by the normal distribution model.

It is concluded that additional creep data are required, for reliable prediction of clad lifetimes in any event, and hopefully for determination of the proper prediction model. These tests should be designed to investigate "creep to leakage" rather than "creep to rupture".

I have recently reviewed the AI document on Pt-20Rh creep, TI-734-250-004, entitled "Creep and Rupture of Pt-20% Rh Pressure Vessels". The absence of this long-awaited document has held the problem of Pt-Rh creep in limbo: discussion was not productive while the prospect of an all-revealing document remained. Now that the document has arrived it is found to be far from all-revealing and further discussion becomes very pertinent. This memo is one result.

Let me make a critical comment at the outset and then get on to more constructive things. I find the report very disappointing. It is my feeling that the program was poorly planned and that the report is also poorly written. More useful data could have been generated than was generated with the effort expended and the report fails to provide a great deal of supporting information that must exist, and which should be supplied for proper interpretation of the test results.

In addition to the above objections, the AI study does not provide an adequate data base for the design of heat sources. Let us first tabulate and analyze the data it does provide.

Six test specimens were employed. These were Pioneer Pt-20Rh clads modified by the addition of a pressurization tube at one end and by welding over the rupture disc in the opposite end. The basic test procedure was to heat the capsules in vacuum by radiation and to load them by internal helium pressure. Diametral strain is measured on two diameters, and onset of leaking is determined by loss of vacuum in the furnace. Details of the six tests are given in Table 1. Tests A-1 and A-2 were intended to relate load to strain rate in the range of interest. Test A-3 was verification of their ability to achieve a desired strain rate. Test A-4 confirmed A-3 and investigated the effect of a reentry temperature pulse. Tests B-1 and

Table 1
Test Plan

<u>Sample Number</u>	<u>Sample Condition</u>	<u>Temperature</u>	<u>Strain Rate, Load Conditions</u>
A-1	no coating no reentry shield	1500°F	10^{-3} to 10^{-2} %/hr variable pressure
A-2	no coating no reentry shield	1500°F	10^{-4} to 10^{-3} %/hr variable pressure
A-3	no coating no reentry shield	1500°F	2×10^{-3} %/hr constant pressure
A-4	no coating no reentry shield	1500°F transient to 2900°F & back	to 1% strain at 2×10^{-3} %/hr pressure reduced
B-1	platinized alumina coating pyrolytic graphite reentry shield	1500°F transient to 2900°F & back	to touch the shield, appr. 2×10^{-3} % strain at 2×10^{-3} %/hr pressure reduced
B-2	platinized alumina coating copper-ZrO ₂ reentry shield	1500°F transient to 2900°F & back	to touch the shield appr. 2×10^{-3} % strain at 2×10^{-3} %/hr pressure reduced

B-2 repeated A-4 under conditions of potential contamination from an emissive coating and reentry heat shield. Strain was measured continuously by an optical cathetometer on tests A-1 through A-4. View of the capsule was obscured by the heat shield in tests B-1 and B-2. Contact of the capsule with the heat shield was determined by electrical contact points.

Test results are shown in Table 2; a number of points are worthy of note. First, it is confirmed that when failure occurs as a result of slow creep deformation it occurs via an intergranular crack or microcrack and results in a slow non-violent release of the pressure. This type of failure occurred in all cases except sample A-4 and in this case the failure occurred during a temperature spike to 2770⁰F when the strain rate was undoubtedly quite high. Sample B-1 also failed during a temperature spike but with the sample supported by a graphite sleeve and unable to deform rapidly the failure was via a slow leak. This type of behavior is to be expected when deformation is by creep, but confirmation is valuable because an energetic release of the pressure would probably cause other damage to the RTG, and it is significant that this is unlikely to happen.

The second point worth of comment is the considerable scatter in the failure points, whether defined in terms of strain, strain rate, or time at temperature. Listing all failures that occurred at 1500⁰F, we have the data shown below in Table 3.

Table 2
Test Results

	<u>Temp</u> oF	<u>Pressure</u> psi	Nominal ⁽¹⁾ <u>Stress</u> psi	Average ⁽²⁾ <u>Strain Rate</u> %/hr	Cumulative <u>Time</u> hrs	Cumulative ⁽³⁾ <u>Strain</u> %	<u>Remarks</u>
A-1	1500	57.7→148.4	3960→10,200	4.3×10^{-3}	150	0.7 (f)	weld leak repaired, test continued
	1500	148.4→168.5	10,200→ 11,550	3.1×10^{-2}	190	2.1 (f)	intergranular leak in cylinder. sample fine-grained
A-2	1500	45→115	3080→7900	4.5×10^{-4}	2600	1.2	large change in strain rate.
	1500	115→140	7900→9600	8×10^{-3}	3250	7.45 (f)	details of failure not described. sample coarse-grained
A-3	1500	88.2	6050	1.7×10^{-3}	3835	7.8	uniform strain rate did not fail sample fine grained
A-4	1500	88.2	6050	2.0×10^{-3}	695	1.22	uniform strain rate temp. pulse begun
	to 2770	45→60	3080→4100	n. a.	n. a.	n. a. (f)	capsule burst, large intergranular tear, sample coarse grained
B-1	1500	88.2	6050	n. a.	647	n. a. (f) est. 1.25	leak in weld heat zone repaired, test continued
	1500	88.2	6050	n. a.	1049	2.25	graphite contact test continued
	1500	88.2	6050	n. a.	2129	n. a.	temp. pulse begun
	to 2675	45→?	3080→?	n. a.	+10.5 min.	n. a. (f) est 2.25+	v. slow leak near weld
B-2	1500	88.2	6050	n. a.	1683	n. a. est 2.5	copper ZrO ₂ contact test continued.
	1500	88.2	6050	n. a.	2210	n. a. est 3.2	slow leak, center of capsule

Notes to Table 2

(1) Nominal stress is defined as:

$$\text{Stress} = \frac{P \times 2.468}{2 \times .018} \text{ where } P = \text{pressure}$$

2.468 = capsule diameter
.018 = capsule wall thickness

However, the manufacturing tolerances on wall thickness are +.002, -.001, a total variation of 16.7%. Since no information is provided on the actual wall thicknesses, the stresses may vary by an equal amount.

(2) Average strain rates are measured from straight lines drawn between the points of major slope change.

(3) Within this column the symbol (f) indicates that a failure, i.e., a leak has occurred. For sample B-1, estimates of strain were taken from the curve for sample A-3 which is particularly well behaved. For sample B-2, estimates of strain were calculated from capsule and shield dimensions provided in the report.

Table 3
1500°F Failures

	Nominal <u>Stress</u> psi	<u>Strain Rate</u> %/hr	<u>Time</u> hr	<u>Strain</u> %	<u>Location of leak</u>
A-1	10.200	4.3×10^{-3}	150	0.7	weld
A-1	11.550	3.1×10^{-2}	190	2.1	cylindrical wall
A-2	9.600	8×10^{-3}	3250	7.45	cylindrical wall (est) (2)
B-1	6050	2×10^{-3} (est)	647	1.25 (est)	weld heat zone
B-2	6050	2×10^{-3} (est)	2210	3.2 (est)	cylindrical wall
A-3 ⁽¹⁾	6050	1.7×10^{-3}	>3835	>7.8	unknown

(1) to increase the data base we may include A-3, noting that conditions to failure must be somewhat better than those at the test termination.

(2) the symbol (est) indicates that the point of information is not provided in the report but may be inferred from related data.

It might be argued that the first and fourth failures in the table should be eliminated from consideration because they were associated with the welds. Although the AI report does not discuss the rationale for their actions, the fact that they repaired these failures and continued the tests suggests that they considered these failures to atypical. An argument will now be presented that all six values should be used in calculations of capsule lifetime.

The first A-1 failure at 0.7% strain was definitely in the cap-to-body weld. Since all real capsules will contain such welds, failure within a weld is a valid possibility and the failure that did occur should not be ignored

except for cause. The report provides no information on how the weld was formed and none on what the weld structure looked like. In the absence of proof that the weld was improperly formed, we see no justification for ignoring a real failure.

There is even less justification for excluding the first B-1 failure at 1.25% strain. This failure occurred in the heat-affected zone of a similar weld. Even if the weld itself were improperly formed, the heat-affected zone is unlikely to differ markedly from that in a real capsule clad. In the absence of hard evidence to the contrary, this failure should be considered to be a real example of a credible failure mode.

The procedure of repairing a first leak and returning the sample to test may be an acceptable or even desirable procedure for exploring the population of all potential failures, providing all failures are considered as equally real. A mechanistic discussion of the creep process is presented below, which argues that the formation of a leak in a thin walled sample deforming in creep is a highly random process dependent upon the grain structure in the sample. Identification of not only the weakest spot but also the second weakest and perhaps even the third weakest, may be an efficient method of exploring the life capabilities of the capsules.

It should be noted that the six data points presented above in Table 3 differ from the four data points that have been used by Teledyne Isotopes and attributed to the AI report. Isotopes has used the values 2.0, 7.0, 7.5 and 8.0 % elongation for their calculations of clad lifetimes. In telephone conversations they have indicated that these numbers were obtained from AI in several different communications, verbal and written. It appears that $2.0 = 2.1$, $7.5 = 7.45$, and $8.0 = 7.8$. However, there is no real point corresponding to 7.0. One can guess that the figure 7.45 got rounded up to 7.5 in one communication and down to 7.0 in another. Elimination of the 7.0 figure would probably not change Isotopes results a great deal although it would represent a large reduction in the size of their data base

and hence in the confidence in their result. Use of all six numbers, as is argued here to be correct, would sharply degrade their reliability prediction. Elimination of the 0.7% point because of questionable weld quality and use of the other five points would still reduce their prediction because of the greater scatter in the larger set of data.

We pass now from discussion of the AI data to presentation of an hypothesis of the mechanism involved in creating a leak through creep.

To appreciate the distinction between deformation at high strain rates and creep, it is necessary to consider how the mechanism of deformation changes from one case to the other. The deformation of metals in response to applied loads is accomplished by the motion of dislocation lines through the individual grains of the metal. Dislocations are, of course, defect structures of the crystals. The strength of a metal is determined in large part by the presence of foreign particles, defects, grain boundaries, etc. that interfere with the motion of dislocations. In a conventional room temperature deformation test the sample deforms at a high strain rate under an increasing load until it breaks. As dislocations encounter obstructions they are blocked for a time until the rising stress is sufficient to force them through or around the obstacle. In general, if the stress is held constant at a level below the breaking stress, the dislocations will remain immobile and the metal will sustain the applied stress indefinitely. In creep, the load is characteristically constant and the metal continuously deforms at a low strain rate. The difference in behavior arises from the fact that creep occurs at an elevated temperature where diffusion at a useful rate is possible. Dislocations are, by their structure, constrained to move in a single crystallographic plane; when an obstruction exists in that plane they are blocked at low temperatures. When diffusion is present it is possible for a dislocation to move out of its crystallographic plane by absorbing or giving up atoms to the crystal around it, thereby passing over or under the obstruction. The diffusion process generates vacancies within the crystal.

These vacancies diffuse in turn to the nearest sink, usually a grain boundary. Those grain boundaries that initially were high-energy boundaries and that are located perpendicularly to the principal tensile stress are the most efficient sinks for vacancies and collect vacancies until they separate and create a void. Failure ultimately occurs when sufficient voids have formed to raise the effective stress level, and accelerate deformation.

The point of relevancy to the present problem is that creep generates vacancies, vacancies condense into voids, and voids link up to form a leak path. With a thin walled gas containment system, failure by leaking can occur at much lower deformations than fracture of the sample. The occurrence of a leak probably depends on the chance that a sufficient number of high-energy grain boundaries exist in sufficiently close proximity to link up and form a leak path. This is a random event; occurrence of leaks should then exhibit a high degree of scatter. The total strain in creep is the proper parameter to predict the occurrence of a leak. The metallurgical structure of the capsule may exhibit a strong influence, however.

Even granting the validity of the above argument, it might be thought that capsules of the size tested contain such a large number of grains that all possible arrangements would exist in each sample, which should lead to reproducibility of strain-to-leak. A brief analysis of the situation will show that this is not so and will also lead to some ideas of what the leak sites probably are. Photomicrographs of samples A-1 and A-3 after failure are provided by AI. They are defined as fine grained. The grains are seen to be fairly uniform in size, equiaxed, and to have average diameters of the order of 0.001 inch. The cylindrical surface of the test capsule has a diameter of 2.468 inch and a length of 2.329 inch, for a total area of 18 in^2 . Since the area of a single grain is 10^{-6} in^2 , there are 1.8×10^7 grains in the surface of the cylindrical portion of the capsule. Since the wall thickness is 0.018 inch, there are 18 grains across any wall thickness. Considering the incidence of voids in a specimen after creep deformation, as shown in Figures 19 & 21 in the AI report,

it appears that something like one grain in five has an associated void. Thus the probability that any particular grain has the proper orientation and grain boundary energy to develop a void is 0.2. Taking the naive position that a leak consists of a chain of 18 such grains lined up across the wall thickness, the probability that any individual grain, visible at the surface, constitutes the end of a leak chain is $(0.2)^{18}$ or approximately 10^{-12} . Since there are only 1.8×10^7 such grains in a sample, the probability of a leak by this process is very small. Put another way, it could be said that the samples tested are really much too small for each one to contain a minimum energy potential leak path, hence the scatter in results.

From this argument one can conclude that the real leak paths probably occur where there are a few large grains of proper orientation within the wall thickness. This would reduce the exponent in the expression for a leak probability and permit a significant number of potential leak paths to exist in a real capsule. This argument also suggests that the heat affected zone near the weld is a likely location for leaks because of local grain coarsening.

The data provided by the AI report are too few, and the test conditions too varied and ill defined, to be of any use in proving or disproving the above hypothesis. Nor are they adequate to provide a sound basis for an empirical calculation of capsule life capability. On the basis of the existing data it is possible to reach the following conclusions:

1. If failure occurs by creep at constant temperature it will be a "soft" failure in which the gas pressure is released non-destructively.

2. Total strain is the parameter to be controlled in design. Allowable total strain for gas containment with high reliability cannot be calculated accurately but evidently is quite low, probably below one percent.

[Using the six values of strain to failure in Table 3, the following may be calculated:

$$\bar{\epsilon} = 3.78\%$$

$$\sigma = 2.86\%$$

$$\sigma' = 3.13\%$$

From these values, it can be shown that the strain that yields a 75% probability of survival with a 75% confidence limit is 0.32%]

3. Additional creep testing is needed

The mechanism proposed above suggests the following additional conclusions:

4. There is probably a limit of strain below which a leak will never develop because insufficient vacancies have been generated to create a leak path even in the most likely leak path. That path would be a single grain boundary of the worst energy and orientation extending the full wall thickness. The calculations reported under conclusion 2 above are based on the assumption of a normal distribution of strains to failure and does not include the concept of a critical strain limit. If the mechanism of random leak paths is correct, then the reliability model based on a normal distribution is not the correct one. The binomial model should be used and the results might be quite different.

5. Use of a close fitting graphite sleeve that limits the possible strain of the clad is beneficial but it must be quite close-fitting. If significant creep occurs, with the attendant generation of void volume, the possibility exists that rearrangement of the voids through sintering, grain growth, etc. might result in a leak developing even after deformation has been stopped by the graphite. However, this would be quite slow: lifetime before failure by this mechanism could be acceptable.

6. If additional creep testing is performed it should be done by a gas pressurization technique. The samples need not duplicate the capsule clad in geometry but should duplicate it in wall thickness and in all metallurgical parameters, including the presence of a production quality weld. The technique of repairing the first leak and continuing the test should be considered as a possible economy technique. However, careful metallurgical examination should be performed on all failures which may preclude their reuse. Use of conventional creep-to-rupture tests would not provide useful information relative to the current problem of creep to leakage.