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SNAP 8 Experimental Reactor Fuel Element Behavior:
Atomics International Task Force Review

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SNAP 8 EXPERIMENTAL REACTOR
FUEL ELEMENT BEHAVIOR:
ATOMICS INTERNATIONAL
TASK FORCE REVIEW*
(Title Unclassified)

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*The work of the Task Force, as reported in this Memo, was completed in June 1966. The Memo does not reflect data developed subsequent to that time.

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ABSTRACT

The Task Force scope was to review, evaluate, and make recommendations on Atomic International postirradiation examination of SNAP 8 Experimental Reactor (S8ER) fuel elements. Emphasis was on density changes, dimensional changes, hydrogen composition, and microstructure.

Evaluation: Irradiation conditions were extended well beyond previous experience, on which preliminary correlation was based; so that S8ER element dimensional behavior could not be called anomalous. The primary cause of cladding cracking was fuel swelling. At least some of the swelling was anisotropic. Hydrogen loss from cracked elements was greater than that from intact elements, by a factor of not more than two, suggesting the presence of an effective barrier, formed on the fuel rod in cracked elements. Fuel swelling was affected by hydrogen loss and redistribution (which resulted in some superhydriding at the cold ends of fuel rods), by phase changes in both the $\delta \rightarrow \beta + \delta$ and $\delta \rightarrow \delta + \epsilon$ regions, and possibly by additive content, as well as by fission product agglomeration, including fission gas bubbles in the hydride matrix. Fundamental fission effects in this complex fuel system have not been worked out; so that the only practicable correlation, at present, must be based on empirical treatment of data.

Recommendations: These were made to AI project personnel, from time to time, in the course of the Task Force work. They included electron microscopy of replicas of irradiated fuel, x-ray diffractometry of unirradiated fuel to supplement metallographic identification of phases in the Zr-H system, and dilatometry of the $\delta \rightarrow \delta + \epsilon$ region in unirradiated fuel to determine volume changes from hydrogen composition changes and thermal cycling. Recommendation for future work included irradiation testing of the effect of additive content in small increments around the 0.2 wt % level, irradiation testing of unfueled zirconium hydride, and the use of x-ray diffractometry for phase identification of irradiated fuel.

Other Findings: A revised phase diagram for the Zr-H system was prepared. Also, analytical expressions were devised for the variation in hydride density as a function of H/Zr; and a systematic analytical procedure was developed for converting observed fuel density changes into design values for diametral and radial fuel-cladding gaps.

TABLE I-1
S8ER FUEL ELEMENTS EXAMINED IN DETAIL

Element	Core Position	Initial H/Zr _(eff) [*]	Carbon (wt %)	Postirradiation Condition	
				Cladding	Fuel
111	IX-42	1.75	0.01	Cracked§ (8 to 13 in.)	Intact
188	VIII-6	1.76	0.0	Intact	Broken
206	IX-15	1.75	0.48	Intact	Intact
232	VI-24	1.73	0.54	Intact	Broken
293	IX-29	1.74	0.0	Intact	Broken
302 [†]	IX-5	1.74	0.50	Intact	Broken
309	V-5	1.69	0.48	Cracked (4 to 9-1/2 in.)	Broken
332	VIII-25	1.73	0.55	Weld Failure	Severely Dehydrated
342	I-1	1.72	0.37	Cracked (2 to 9 in.)	Intact
378	IX-28	1.69	0.34	Cracked (4 to 12 in.)	Broken
398	IV-13	1.70	0.40	Cracked (4 to 8 in.)	Severely Dehydrated
432	VIII-29	1.74	0.54	Intact	Intact
474	II-4	1.80	0.49	Intact	Intact
476	VII-19	1.79	0.50	Cracked (4 to 9-1/2 in.)	Broken
486 [†]	III-3	1.77	0.19	Cracked (6-1/2 to 11 in.)	Not Examined
519	IV-1	1.72	0.50	Cracked (5-1/2 to 10-1/2 in.)	Broken
525	VII-28	1.69	0.31	Intact	Intact

*H/Zr, in this report, indicates an "effective" hydrogen-to-zirconium ratio, calculated on the basis that the zirconium (added to the fuel as ZrC) does not hydride.

$$H/Zr_{(eff)} = \frac{90.496 \text{ (wt \% H)}}{100 - 8.595 \text{ (wt \% C)} - \text{(wt \% U)} - \text{(wt \% H)}}$$

[†]Only limited postirradiation examination

§Crack locations indicated by distance from top of element

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I. INTRODUCTION AND SUMMARY

By
H. Pearlman

This report presents the findings of a Task Force that reviewed the Atomic International effort on SNAP 8 Experimental Reactor (S8ER) fuel element evaluation, over the interval from February to June 1966. The purpose and composition of the task force are contained in the Appendix to this report.

At the time the task force was established, postirradiation examination of the S8ER core had revealed that all 211 elements were substantially deformed and bowed, and the cladding of 167 elements had cracked. Detailed examination of a group of elements with both cracked and intact cladding had begun. This included metallography and microhardness, density, hydrogen content, and burnup determinations at selected locations in the fuel rods, and determination of the stress-strain characteristics of the cladding of another group of intact elements. At the end of the time period during which the Task Force was functioning, the detailed fuel element examination was essentially complete, as were also the postirradiation examinations of Capsule Experiments NAA 82-1 and NAA 115-1. During the time that these examinations were being conducted, several reports were published on results of earlier phases of the S8ER investigation, and many results of the detailed examinations were incorporated in briefing documents. (I-1, -2, -3)

Table I-1 lists the S8ER elements in the detailed examination program, along with some of their characteristics. (I-4) These elements were selected on the basis that they had been representative of different families:

- 1) Cracked or intact
- 2) Maximum distortion at top, middle, or bottom
- 3) High, normal, or low gamma activity
- 4) Core position.

It is concluded that the basis for selection was sound. In addition to these elements, five others were sectioned perpendicular to the axis and studied

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TABLE I-2
ELEMENTS STUDIED IN
CROSS SECTION ONLY

Element	Core Position	Condition
351	IV-12	Cracked
388	VI-25	Intact
410	VII-27	Intact
492	III-2	Cracked
519	IV-1	Cracked

metallographically to provide detailed information about the fuel-barrier-cladding region, as well as about the fuel itself. The cladding was not removed from these elements, which are listed in Table I-2.

We followed closely the examination of the S8ER elements and of the NAA 115-1 and 82-1 capsules, and also reviewed earlier fuel irradiation data and S8ER operational history.

With regard to the S8ER fuel swelling and cladding cracking, our general conclusion is that these results are not "anomalous." That is, there was no set of fuel irradiation data, prior to the start of S8ER examination, that provided an adequate frame of reference against which S8ER fuel performance could be judged. A summary of detailed conclusions follows.

A. CLADDING CRACKING IN S8ER

The cracking in the S8ER cladding was caused by fuel deformation ("swelling") that stressed the Hastelloy N cladding.

B. NATURE OF FUEL DEFORMATION - SWELLING, $\delta \rightarrow \beta + \delta$ TRANSITION

Analysis of capsule experiment data, as well as S8ER elements, indicates that fuel swelling alone, without the superimposed phenomena associated with metallurgical phase changes in the hydride, could have stressed the cladding to failure. Small clearances between the fuel and the barrier/cladding, especially those in the axial direction which limited axial growth early in life, led to stresses that contributed to the cladding deformation and cracking.

There is no question that some of the elements operated partially in the β phase. A post-test H/Zr ratio of <1.33 , or a microstructure characteristic of decomposed eutectoid, is evidence of such operation. The $\delta \rightarrow \beta$ transition is accompanied by a decrease in specific volume; in other words, the fuel shrinks. However, the predominant mode of cladding cracking implies a hoop

stress. Thus, other concomitant mechanisms must have been operating to cause the fuel to swell to produce such a stress. These may be: (1) fission gas bubble growth or cavitation, induced by the phase change, and/or (2) the radially outward deformation of the mechanically weaker β -phase material, under strong compressive loading associated with the axial restraint (see Section VII).

C. METALLURGICAL PHASE IDENTIFICATION - $\delta \rightarrow \beta$ REGION

Postirradiation optical microscopy does not provide definite indication of the phases present, particularly in the H/Zr range near 1.40, where the β structure may have decomposed to $\alpha + \delta$.

X-ray diffraction studies on unirradiated material of comparable H/Zr show no α phase present, at or above H/Zr of ~ 1.44 . This and other facts suggest that another phase, here designated γ , is stable at H/Zr ratios > 1 , at temperatures to $\sim 250^\circ\text{C}$ (490°F). A modified phase diagram is presented to account for the observations on SNAP 8 fuel (see Section II).

D. PHENOMENA AT COLD ENDS OF FUEL RODS - $\delta \rightarrow \epsilon$ REGION

At the fuel temperatures prevailing near the coolant inlet end of the core, the effects of superhydriding arising from hydrogen redistribution impose a different swelling mode. This is associated with the $\delta \rightarrow \epsilon$ phase transition, and/or the increased H/Zr ratios in pre-existing ϵ . The anisotropy of the tetragonal crystal structure of the ϵ phase, and the decreasing density with increasing H/Zr, could provide mechanisms for fuel swelling in this region (see Section VII).

E. CORRELATION OF SNAP 8 FUEL SWELLING DATA

Development and application of an empirical correlation, based on capsule irradiation data, to S8ER elements or other elements operating in a reactor core is made difficult by at least three factors:

- 1) Hydrogen redistribution caused by temperature gradients, especially axial gradients
- 2) The value of the temperature chosen for use in the correlation. The surface temperature is more representative of the fuel conditions, at a given axial location, than is the centerline temperature; the best value is an average.

3) Theoretical considerations, based on fundamental fission effects in the dispersed uranium-zirconium hydride fuel system, are not sufficiently worked out to offer guidance for a correlation.

The correlation developed and reported in NAA-SR-9633 is principally of historical interest. An updated representation of swelling data seems to require a term for fuel composition, as well as terms for temperature and burnup. An approach to such a correlation is presented in Section VI.

F. ANISOTROPY OF VOLUME CHANGES IN SNAP 8 FUEL

The observed density (volume) changes may be corrected for hydrogen loss by application of an appropriate factor. The distribution of the volume correction, to diameter and to axial length, is not isotropic. In the corrected volume change (assumed independent of the hydrogen loss phenomenon, and assigned to burnup effects), the distribution of the change is also not isotropic. The degree of burnup anisotropy appears to depend upon the carbon content (see Sections III and VI).

G. HYDROGEN DISTRIBUTION IN FUEL ELEMENTS

H/Zr profiles for both cracked and intact elements in the detailed examination show that the greatest hydrogen losses occur nearer to the coolant outlet (top) ends of the elements than at the maximum centerline temperature location. It is conceivable that redistribution of hydrogen, during the interval of 1200°F outlet temperature operation before reactor shutdown, could have caused some shifting of the minimum H/Zr values. It is not surprising, however, that the lowest H/Zr values appear so close to the top ends of the elements, because the hydrogen distribution would be expected to be more dependent on surface or radially averaged fuel temperature than on centerline temperature.

Integration of plots of $\Delta(H/Zr)$ vs axial length (where Δ is the initial value minus the final value) indicate that the mean hydrogen loss in the cracked elements was perhaps no more than a factor of 2 greater than that in the intact elements. It is postulated that a surface barrier, formed in the cracked fuel because of the oxidizing environment locally present, effectively prevented greater loss (see Section IV).

H. THERMOMECHANICAL EFFECTS

Stresses imposed on the cladding as a result of thermomechanical effects, even those accompanying reactor scram, appear to be too small to constitute the primary cause of cracking.

I. RECOMMENDATIONS

The following work is recommended in further evaluation of the S8ER elements.

1. H/Zr Profiles in S8ER Fuel Rods

Measurement of the H/Zr axial profiles in several more elements should be performed, with emphasis on establishing the location of the minimum H/Zr values (see Section V).

2. Postirradiation Annealing of Irradiated Fuel

Two types of fuel annealing experiments should be run. In one (isothermal annealing, under conditions where the sample does not lose hydrogen), the results will indicate the possibility of annealing microcracks and also provide some information on agglomeration of fission product gases. In the other, an irradiated sample is sealed in a hydrogen-tight container with unfueled hydride, which will either provide hydrogen to the sample or remove hydrogen from it. This will permit cycling through the $\beta \rightarrow \beta + \delta$ region, or vice versa; and the results should indicate the influence of metallurgical phase transition on the agglomeration of fission gases.

It also would be of interest to anneal sections of cladding with attached barrier layer, to provide elucidation of fission fragment entrapment in the barrier. Annealing at a sufficiently high temperature should release trapped fission gases (see Section VII).

3. Phase Identification in Dehydrated Fuel, $\delta \rightarrow \beta$ Region

Using unirradiated fuel, dehydrated and then rehydrated under controlled conditions, metallographic examination supplemented by x-ray diffraction should provide clarification of the phases presented in irradiated material at known H/Zr (see Section I-C).

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4. Volume Changes in $\delta \rightarrow \epsilon$ Region

Unirradiated fuel, in the H/Zr range of 1.6 to 1.8, should be thermally cycled from room temperature to $\sim 1550^{\circ}\text{F}$, and volume changes followed with a dilatometer. Controlled hydriding at 1200 to 1300°F should be performed, followed by dehydriding, and similarly followed. The results should clarify the fuel phenomena observed at the bottom (inlet) ends of the S8ER elements (see Section I-D).

5. Electron Microscopy of Irradiated Fuel

Examination of replicas of irradiated fuel should be made at $\sim 4000\times$ or higher. The microstructural phenomena associated with operational effects, such as fission gas bubbles or microcracks, are expected to be too small to be resolved by optical microscopy (see Section VII).

6. X-ray Diffraction Examination of Irradiated SNAP Fuel

Because of the prominence of phase change in determining the behavior of SNAP fuel, it is highly desirable to acquire the capability of performing x-ray diffraction examination of irradiated material.

7. Influence of Carbon Content

In addition, future irradiation experiments should be considered which explore the possible dependence of swelling upon fuel composition, using smaller increments of carbon content than in past irradiations (see Section VI).

8. Irradiation of Unfueled Zirconium Hydride

Also suggested are irradiation experiments on unfueled zirconium hydride (see Section VII).

The following sections of this report consist of selected topics, developed by task force members during the review, that have application to SNAP fuel technology in general, as well as to the S8ER fuel.

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- I-3 SNAP 8 Briefing, Washington, D.C., May 17, 1966, AI-66-Memo-99 (This is the most recent of several briefing documents)
- I-4 Data in Table I-1 are taken from the series of progress letters from the SNAP 8 project to AEC. A listing of the more recent letters is: 66AT-2970 (May 25, 1966), 66AT-2600 (May 11, 1966), 66AT-2254 (April 27, 1966), 66AT-1890 (April 13, 1966), 66AT-1732 (March 30, 1966), 66AT-1358 (March 22, 1966), 66AT-1210 (March 2, 1966)

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Figure II-1. Dissociation Pressures in the Zr-H System

Figure II-2. Phase Diagram of Zr-H System

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II. PROPOSED PHASE DIAGRAM OF THE Zr-H SYSTEM

By
J. B. Vetrano

The earliest recorded investigation of the zirconium-hydrogen system dates back to Hägg's x-ray study in 1929. During the intervening years, many investigators have attempted to clarify and expand on Hägg's original work. They have used a variety of methods for investigating phase boundaries and phase transformations. These have included metallography, dilatometry, electrical resistivity, Hall coefficient, internal friction, heat capacity, thermal diffusivity, and dissociation pressure measurements. It is this same variety of methods which has added confusion, as well as clarification, to the proper positioning of the proposed phase boundaries. In many cases, an abrupt change in one property may appear as a smooth transition in another, leaving doubt as to whether a phase boundary has been crossed. Furthermore, disagreement exists over the issue of whether two of the phases observed (γ and ϵ) are stable or metastable.

Through all of these investigations comes a certain consistency, however. All those who have investigated the Zr-H system agree that it is undoubtedly a eutectoid-type system, with hydrogen serving to stabilize α zirconium to temperatures as low as 550°C, the eutectoid temperature. There is also general agreement that, between H/Zr atom ratios of about 1.6 and 2.0, there exists one or more hydride compounds, possessing neither the structure of α or β zirconium. Beyond this, for every single specific feature of the diagram proposed by any investigator, there is some contradictory evidence. Two representations which have been useful in the engineering application of zirconium hydride to the SNAP reactor concepts are shown in Figure II-1, which is a P vs T plot, and in Figure II-2, which is a T vs H/Zr plot. Figure II-1 avoids the issue of fine structure in the phase diagram, and is used primarily for determining dissociation pressures under various conditions. In Figure II-2, the temperature and composition limits of the various phases are shown, the dashed lines indicating considerable uncertainty.

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Figure II-2 shows the lowering of the α - β transformation temperature of Zr by the addition of hydrogen, the limited solubility of hydrogen in α zirconium, the extensive solubility of hydrogen in β zirconium, and the eventual formation of a hydride compound phase existing over a fairly broad stoichiometry range. The principal points of uncertainty are:

- 1) Definition of the composition limit of the δ -hydride phase below the eutectoid temperature
- 2) Accommodation of the frequently observed γ -phase in the equilibrium diagram
- 3) Definition of the δ - ϵ transformation.

Although the accommodation of a γ structure in the equilibrium diagram is quite controversial, we prefer to show it as a stoichiometric compound, $\text{ZrH}_{1.0}$, existing at temperatures up to $\sim 250^\circ\text{C}$. This representation is based, primarily, on room temperature metallography of unirradiated zirconium hydride. Because of the emphasis placed on metallography in the postirradiation examination of the S8ER, this form of the diagram is particularly valuable for explaining S8ER fuel behavior.

The δ - ϵ boundary is shown as a miscibility gap, closing up below 400°C . The two-phase region at room temperature is based on metallography and x-ray diffraction work by numerous investigators. The continuous transformation from cubic δ to tetragonal ϵ at elevated temperatures is based on dissociation pressure studies by numerous investigators and on limited x-ray diffraction studies at 400°C by Motz.

The phase diagram proposed here has several features of that of Raymond. Observations on S8ER fuel, where the phase identified as γ' was frequently encountered, have strengthened the idea that the phase - here, called γ - may be stable over a limited temperature range.

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III. ANALYSIS OF DIMENSIONAL CHANGES IN HYDRIDE FUEL

By
C. K. Smith

In hydride fuels, the total volume changes observed during irradiation are attributable to two separate factors:

- 1) Fission swelling
- 2) Changes in hydrogen content, occurring as a result of both hydrogen loss from the fuel and hydrogen redistribution in the fuel.

Because the relative magnitude of these factors is a function of many variables, any attempt to understand the behavior of hydride fuels requires study of the volume change attributable to each alone.

This section presents the equations for making the necessary interpretation of the experimental observations. Nomenclature used in the analyses are presented at the end of the section.

A. VOLUME CHANGES

1. General Relationships

The pre- and postirradiation data available for computation of volume changes are obtained from measurements of densities and hydrogen contents. From the definitions

$$V = \frac{W}{\rho}$$

and

$$\Delta(\text{Quantity}) = (\text{Quantity})_{\text{final}} - (\text{Quantity})_{\text{initial}} ,$$

the volume change is

$$\Delta V = V_f - V_i = \frac{W_f}{\rho_f} - \frac{W_i}{\rho_i} = \frac{W_i + \Delta W}{\rho_i + \Delta \rho} - \frac{W_i}{\rho_i} .$$

Dividing by

$$V_i = \frac{W_i}{\rho_i}$$

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yields

$$\frac{\Delta V}{V_i} = \frac{\rho_i}{\rho_i + \Delta \rho} \times \frac{\Delta W}{W_i} - \frac{\Delta \rho}{\rho_i + \Delta \rho} \quad \dots (III-1)$$

2. Density Correction Due to Change in Hydrogen Content

Examination of the density curves (Figure III-1) for hydride fuels as a function of $H/Zr_{(eff)}$ indicates that, over the range of uranium contents of interest here, the density change due to changing hydrogen content is given by:

$$(\Delta \rho)_h = \begin{cases} 0.0975 (R_i - R_f) & 1.59 \leq R_i \text{ AND } 1.59 \leq R_f \\ \left. \begin{array}{l} 0.393 + 0.0975 R_i - 0.345 R_f \text{ (LOWER LINE)} \\ 0.685 + 0.0975 R_i - 0.528 R_f \text{ (UPPER LINE)} \end{array} \right\} & 1.59 \leq R_i \text{ AND } 1.30 \leq R_f \leq 1.59 \\ \left. \begin{array}{l} 0.685 + 0.0975 R_i - 0.569 R_f \text{ (LOWER LINE)} \\ 0.685 + 0.0975 R_i - 0.528 R_f \text{ (UPPER LINE)} \end{array} \right\} & 1.59 \leq R_i \text{ AND } 0 \leq R_f \leq 1.30 \end{cases} \quad \dots (III-2)$$

where R is the ratio $H/Zr_{(eff)}$.

Values of $(\Delta \rho)_t$ are obtained from direct measurements of ρ_f and ρ_i , and values of $(\Delta \rho)_h$ are calculated from Equation III-2. The volume change, or density change, assigned to burnup alone may be calculated using the definitions

$$\left. \begin{array}{l} (\Delta V)_{bu} = (\Delta V)_t - (\Delta V)_h \\ (\Delta \rho)_{bu} = (\Delta \rho)_t - (\Delta \rho)_h \end{array} \right\} , \quad \dots (III-3)$$

on the assumption that the individual changes add independently. This provides a means of adjusting experimentally determined $(\Delta \rho)_t$ values to a constant value of R , equal to the initial value. $(\Delta \rho)_{bu}$ derived in this way may then be related to the inherent irradiation performance of the fuel. The $(\Delta \rho)_h$ values take into account the effectiveness of the hydrogen barrier and the influence of phase change. In principle, then, the assumption made permits an estimate of the

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Figure III-1. Density of Hydride Fuel as a Function of Uranium and H/Zr

change in fuel density in the hypothetical condition where no hydrogen is lost. Without the correction through $(\Delta \rho)_h$, it would be impossible to compare the results of one capsule irradiation test with another, or relate the observations on one fuel element to another.

The process that causes the "pure" burnup swelling may interact with the processes of dehydriding and rehydriding. There is evidence that they do so when phase change is involved.^(III-1) As a consequence, it is not apparent, for example, that the $(\Delta \rho)_{bu}$ values calculated for the following two irradiation histories would be equal:

- 1) Burnup to m metal at. % at constant temperature, where hydrogen is lost monotonically with time through the barrier, to some final concentration
- 2) Identical burnup of the same starting material at essentially the same temperature, where some hydrogen migration has occurred, in addition to some loss through the barrier, to yield the same final concentration.

Nevertheless, in the present state of knowledge, the $(\Delta \rho)_h$ correction seems to be the best that can be made.

3. Calculation of Weight Change

The only sources of weight change are burnup and hydrogen loss or gain. Therefore,

$$\frac{(\Delta W)_t}{W_i} = \frac{(\Delta W)_h}{W_i} + \frac{(\Delta W)_{bu}}{W_i} .$$

But the weight changes associated with burnup are negligible, even at relatively high burnups, which means

$$\frac{(\Delta W)_{bu}}{W_i} = 0 , \quad \dots (III-4)$$

and

$$\frac{(\Delta W)_t}{W_i} = \frac{(\Delta W)_h}{W_i} .$$

But

$$\frac{(\Delta W)_h}{W_i} = \frac{\Delta(\text{weight hydrogen})}{W_i} = \frac{[W_i + (\Delta W)_h] (\text{wt } \% \text{ H})_f - W_i (\text{wt } \% \text{ H})_i}{100 W_i},$$

which reduces to

$$\frac{(\Delta W)_t}{W_i} = \frac{(\Delta W)_h}{W_i} = \frac{(\text{wt } \% \text{ H})_f - (\text{wt } \% \text{ H})_i}{100 - (\text{wt } \% \text{ H})_f} \quad \dots (\text{III-5})$$

4. Volume Change Attributable to Burnup Alone

For radiation swelling alone, Equation III-1 becomes

$$\frac{(\Delta V)_{bu}}{V_i} = \frac{(\Delta W)_{bu}}{W_i} - \left[1 + \frac{(\Delta W)_{bu}}{W_i} \right] \left[\frac{(\Delta \rho)_{bu}}{\rho_i + (\Delta \rho)_{bu}} \right].$$

The weight change associated with burnup of the fuel is negligible. Therefore, the preceding equation reduces to

$$\frac{(\Delta V)_{bu}}{V_i} = \frac{- (\Delta \rho)_{bu}}{\rho_i + (\Delta \rho)_{bu}}.$$

Because $(\Delta \rho)_{bu}$ is not directly measurable, if initial and final hydrogen contents differ, it must be calculated using the relationship

$$(\Delta \rho)_{bu} = (\Delta \rho)_t - (\Delta \rho)_h,$$

where the hydrogen correction, $(\Delta \rho)_h$, is determined from the appropriate Equation III-2. Therefore,

$$\frac{(\Delta V)_{bu}}{V_i} = \frac{(\Delta \rho)_t - (\Delta \rho)_h}{\rho_i + (\Delta \rho)_t - (\Delta \rho)_h} \quad \dots (\text{III-6})$$

5. Total Volume Change

From Equation III-1,

$$\frac{(\Delta V)_t}{V_i} = \frac{\rho_i}{\rho_i + (\Delta \rho)_t} \times \frac{(\Delta W)_t}{W_i} - \frac{(\Delta \rho)_t}{\rho_i + (\Delta \rho)_t} .$$

Substitution of Equation III-5 into this expression gives

$$\frac{(\Delta V)_t}{V_i} = \frac{\rho_i}{\rho_i + (\Delta \rho)_t} \left[\frac{(\text{wt } \% \text{ H})_f - (\text{wt } \% \text{ H})_i}{100 - (\text{wt } \% \text{ H})_f} \right] - \frac{(\Delta \rho)_t}{\rho_i + (\Delta \rho)_t} . \quad \dots (\text{III-7})$$

6. Volume Change Attributable to Hydrogen Alone

By definition,

$$\frac{(\Delta V)_h}{V_i} = \frac{(\Delta V)_t}{V_i} - \frac{(\Delta V)_{bu}}{V_i} .$$

Substitution of Equations III-5 and III-6 gives

$$\frac{(\Delta V)_h}{V_i} = \frac{\rho_i}{\rho_i + (\Delta \rho)_t} \left[\frac{(\text{wt } \% \text{ H})_f - (\text{wt } \% \text{ H})_i}{100 - (\text{wt } \% \text{ H})_f} \right] - \frac{(\Delta \rho)_h}{\rho_i + 2(\Delta \rho)_t - (\Delta \rho)_h} . \quad \dots (\text{III-8})$$

It should be noted that, because burnup and hydrogen effects are defined to be directly additive and it was elected earlier to relate $(\Delta V)_{bu}$ to V_i , Equation III-8 is not directly obtainable from the general relationship represented by Equation III-1.

B. LENGTH AND DIAMETER CHANGES

1. General Relationships

From the definitions

$$V = \frac{\pi}{4} D^2 L$$

and

$$\Delta(\text{Quantity}) = (\text{Quantity})_{\text{final}} - (\text{Quantity})_{\text{initial}}$$

it can be shown that

$$\frac{\Delta V}{V_i} = \frac{\Delta D}{D_i}(2 + r) + \left(\frac{\Delta D}{D_i}\right)^2(1 + 2r) + \left(\frac{\Delta D}{D_i}\right)^3 r ,$$

where

$$r = \frac{\Delta L/L_i}{\Delta D/D_i} .$$

For $\Delta V/V_i \leq 10\%$, a range which covers the NAA 82-1, NAA 115-1, and S8ER observed growth, the error introduced by neglecting the second and third order terms in the $\Delta V/V_i$ equation is $<5\%$. Therefore, for analysis of existing hydride fuel data, it is sufficiently accurate to use

$$\frac{\Delta V}{V_i} = \frac{\Delta D}{D_i}(2 + r) \quad \dots (\text{III-9})$$

Plots showing the relationships of $\Delta L/L_i$ and $\Delta D/D_i$ to $\Delta V/V_i$, for values of r ranging from 0 to 3, are shown in Figure III-2.

2. Diameter and Length Changes Due to Change in Hydrogen Content

For hydrogen loss or gain alone, Equation III-9 becomes

$$\frac{(\Delta V)_h}{V_i} = \frac{(\Delta D)_h}{D_i}(2 + r_h) ,$$

where

$$r_h = \frac{(\Delta L)_h/L_i}{(\Delta D)_h/D_i} .$$

The quantity $(\Delta V)_h/V_i$ can be computed from Equation III-8; and, if r_h is known, $(\Delta D)_h/D_i$ and $(\Delta L)_h/L_i$ can be calculated from the preceding equation.

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Figure III-2. Influence of Anisotropy on Axial and Diametrical Growth

Experiments on dehydriding unirradiated S8ER fuel have, to date, yielded data^(III-1) as follows:

R_i	R_f	$\%(\Delta L)_h / L_i$	$\%(\Delta D)_h / D_i$	r_h
1.52	1.20	-0.55	-0.76	0.73
1.44	0.89	-1.1	-1.25	0.88

These results are consistent with the observations made by Espagno and others^(III-2) on hydriding of extruded Zr-U alloy rods and with the Al hydriding experience,^(III-3) both of which indicate r_h to be less than unity. Therefore, it is suggested that a value of 0.8, which is the average of the two values in the preceding table, be used for r_h ; this gives

$$\frac{(\Delta D)_h}{D_i} = \frac{(\Delta V)_h / V_i}{2.8} \quad \dots (III-10)$$

and

$$\frac{(\Delta L)_h}{L_i} = 0.8 \frac{(\Delta D)_h}{D_i} = \frac{0.8}{2.8} \frac{(\Delta V)_h}{V_i} \quad \dots (III-11)$$

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Selection of a possibly better value for r_h must await the results of additional measurements, including rehydrating of the dehydrated specimens.

3. Diameter and Length Changes Due to Burnup

For burnup alone, Equation III-9 becomes

$$\frac{(\Delta V)_{bu}}{V_i} = \frac{(\Delta D)_{bu}}{D_i} (2 + r_{bu}) ,$$

where

$$r_{bu} = \frac{(\Delta L)_{bu}/L_i}{(\Delta D)_{bu}/D_i} .$$

The quantity $(\Delta V)_{bu}/V_i$ can be computed from Equation III-6; and, if r_{bu} is known, $(\Delta D)_{bu}/D_i$ and $(\Delta L)_{bu}/L_i$ can be calculated from the preceding equations.

Because so much of the existing SNAP fuel data contain axial and/or radial external restraint effects, it is difficult to assess a value of r_{bu} for fuel completely without external restraint. Probably the best present data for unrestrained specimens are those resulting from the NAA 115-1 experiment, for which the relevant values are as follows:

TABLE III-1
DATA FROM NAA 115-1 EXPERIMENT

IS No.	Carbon (wt %)	$\frac{\%(\Delta V)_{bu}}{V_i}$ (middle)	$\frac{\%(\Delta D)_{bu}}{D_i}$ (middle)	r_{bu}
2	0.09	2.59 - 2.90	0.77 - 0.88	1.36 - 1.30
3A	0.04	3.61	0.88	2.10
4	0.40	4.28	1.07	2.00
5A	0.04	4.04	1.04	1.88
7	0.11	3.86 - 4.13	1.21 - 1.31	1.10 - 1.15
9	0.08	1.78 - 1.83	0.54 - 0.56	1.30 - 1.27
10	0.12	1.62	0.49	1.31
11	0.12	1.84	0.51	1.61
12	0.12	1.66	0.52	1.19

The burnup volume change data shown are from Table V-1, with hydrogen corrections made to the diameter change data,⁽¹⁻²⁾ using the techniques described earlier in this section. The IS-1A specimen is omitted from this listing because data on its middle section are unavailable; the IS-6 and IS-8 specimens are omitted because it is highly probable they encountered external restraint at some time during the NAA 115-1 irradiation.

Grouping specimens with similar carbon contents yields:

Carbon (wt %)	Number of Specimens	Average r_{bu}
0.04	2	1.99
0.08 - 0.12	6	1.31
0.40	1	2.00

Therefore, it appears that the SNAP 8 fuel is distinctly anisotropic in its radiation growth behavior, such that the % change in length exceeds the % change in diameter, the degree of anisotropy reaching a minimum at some point between 0.04 and 0.40 wt % C. A statistical analysis* of ΔL data for S8ER, the results of which are shown in Figure III-3, offers some support to this conclusion, in that it suggests sharp changes in swelling behavior might occur in the range of 0.15 to 0.20 wt % C, and possibly in the range of 0.00 to 0.15 wt % C. It is concluded that:

- 1) An assumption of isotropic radiation-induced growth ($r_{bu} = 1.00$) for calculation of diametrical growth is probably conservative for all C contents
- 2) An assumption that $r_{bu} = 2.00$ for calculation of radiation-induced growth in the axial direction is probably conservative for $0.04 \leq \text{wt \% C} \leq 0.40$.

The equations for radial and axial growth, using the limiting values of $r_{bu} = 1.00$ for diametrical growth and $r_{bu} = 2.00$ for axial growth, are

$$\frac{(\Delta D)_{bu}}{D_i} \leq \frac{1}{3} \frac{(\Delta V)_{bu}}{V_i} \quad \dots (\text{III-12})$$

*Made by D. S. Burgess

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Figure III-3. ΔL vs Carbon Content for S8ER With 90% Confident Limits of Mean ΔL

and

$$\frac{(\Delta L)_{bu}}{L_i} \leq \frac{1}{2} \frac{(\Delta V)_{bu}}{V_i} \quad \dots (III-13)$$

To apply these equations to a given element size for given temperature and burnup profiles, the element length should be divided into n increments and the incremental $[(\Delta V)_{bu}/V_i]_n$ for each of the n increments estimated.

The quantities $[(\Delta D)_{bu}]_n$ and $[(\Delta L)_{bu}]_n$ for each of the n increments can then be estimated, using Equations III-12 and -13. The diametrical gap allowed in the element should be greater than the maximum $[(\Delta D)_{bu}]_n$ in any increment along the rod, and the axial gap allowed in the element should be greater than the sum of the $[(\Delta L)_{bu}]_n$ for the n rod increments.

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NOMENCLATURE

<u>Symbol</u>	<u>Quantity</u>
D	Fuel rod diameter
L	Fuel rod length
R	$H/Zr_{(eff)}$
r	Ratio $(\Delta L/L_i)/(\Delta D/D_i)$
V	Volume
W	Weight
ρ	Density

Subscripts:

bu	Attributable to burnup
f	Final value
h	Attributable to change in R
i	Initial value
n	Value in n^{th} increment
t	Total value

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IV. HYDROGEN LOSS AND PHASE CHANGES IN S8ER FUEL

By

J. B. Vetrano

A. PHASE CHANGES

A prime interest during the postirradiation examination of the S8ER elements was to establish whether the fuel had ever lost enough hydrogen to exist in the β phase during reactor operation; and, if so, to determine what the effect was on fuel swelling. This is a particularly difficult set of questions to answer, because all that remained for examination was a room temperature structure, from which it is very difficult to deduce just what steady-state conditions were like during reactor operation. The β phase cannot even be quenched in at room temperature, let alone be retained during the relatively long cooldown period of the reactor from its operation with 1300°F coolant temperature. This period includes the last month of operation at 1200°F coolant outlet temperature. During the cooldown, the hydrogen also has a chance to redistribute from its gradient at operating temperatures, so that postirradiation hydrogen analyses give only an approximation of the hydrogen profile at the operating conditions.

In spite of these difficulties, it is still possible to deduce quite a bit of information from the postirradiation examination. The principal data considered for answering the questions relating to phase change were postirradiation hydrogen analysis, density, and microstructure. For the elements that were subjected to very detailed analysis in the hot cell, there are a total of about 40 axial locations for which postirradiation density, hydrogen content, and microstructure are known. From a careful consideration of these data, it is concluded that:

- 1) Most of the core was δ or ϵ phase at time of shutdown
- 2) A fraction of the core operated so close to the $\delta \rightarrow \beta$ boundary that the postirradiation examination could not distinguish whether any β phase had been present during operation
- 3) A few regions of the core definitely were operating in the β phase at the time of shutdown

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TABLE IV-1
SEGMENTS OF S8ER FUEL HAVING A POST-TEST H/Zr RATIO >1.54

Fuel Element	Distance From Top of Fuel Rod* (in.)	H/Zr		Micro-structure	$\frac{(\Delta V)_{bu}}{V_i}$ (%)
		Pre-Irradiation	Post-Irradiation		
111	3-1/2	1.75	1.59	$\delta + \epsilon$	-
	9-1/2	1.75	1.69	ϵ	-
	11-1/2	1.75	1.74	ϵ	-
206	2-1/2	1.75	1.63	$\delta + \epsilon$	0.58
	11	1.75	1.72	$\delta + \epsilon$	1.35
232	1-1/2	1.73	1.57	$\delta + \gamma$	1.36/1.60
	5-3/4	1.73	1.56	$\delta + \epsilon$	3.68/3.82
	9-1/2	1.73	1.68	$\delta + \epsilon$	-
	11	1.73	1.71	ϵ	-
293	4-1/2	1.74	1.69	$\delta + \epsilon$	-
	5-1/2	1.74	1.62	$\delta + \epsilon$	1.15
	12-3/4	1.74	1.78	ϵ	-
309	10-1/2	1.69	1.63	$\delta + \epsilon$	1.53
332	13	1.73	1.59	$\delta + \epsilon$	0.02/0.16
351	4	1.74	1.56	$\delta + \gamma$ rim with ϵ core	-
	13-1/4	1.74	1.64	$\delta + \gamma$ rim with ϵ core	-
378	3	1.69	1.58	$\delta + \epsilon$	4.76
	9-1/2	1.69	1.68	ϵ	-
	11-1/2	1.69	1.72	ϵ	-
388	3-1/2	1.72	1.68	$\delta + \epsilon$	-
432	9	1.74	1.68	$\delta + \epsilon$	1.13
	11	1.74	1.71	ϵ	0.96
474	3-1/2	1.80	1.71	ϵ	2.84
	8	1.80	1.71	ϵ	2.41
	11	1.80	1.77	ϵ	2.09
476	7-1/2	1.79	1.59	$\delta + \epsilon$	0.88
	9-1/2	1.79	1.69	ϵ	1.72

*Top of fuel rod corresponds to outlet end of reactor

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4) The extent of fuel swelling was not relatable, in a simple way, to the hydrogen content of the fuel during operation.

1. Criteria for δ -Phase Operation

Using the approximation that there was no hydrogen redistribution during reactor cooldown, all sections of the core having an H/Zr ratio >1.44 must have operated in the δ or ϵ region at temperatures below 1500°F (See Figure II-2). While this is admittedly an approximation, and thus not rigidly correct, it is considered adequate for the present analysis. Quite severe hydrogen gradients along fuel rod axes were observed during postirradiation examination, so that the reactor cooldown period was far from sufficient for a complete homogenization. To allow for partial homogenization, let us establish the arbitrary criterion that 0.1 H/Zr units could be absorbed from other areas of a fuel rod on cooldown. This then leads to the criterion that any section with a post-test H/Zr ratio >1.54 must have been operating in the δ or ϵ phase at the time of reactor shutdown. All such sections of fuel are listed in Table IV-1, ^(IV-1) along with the observed postirradiation microstructure and density. Also included in this table is the percent volume change corrected for hydrogen loss.

An explanation needs to be made about the microstructure designation in Table IV-1. The presence of any banding at all was used as a criterion for saying that ϵ phase was present. Actually, in many of the specimens, particularly those with lesser amounts of hydrogen, the banding was quite indistinct. It is quite possible that, in these cases, the material was really dehydrided ϵ (i. e., δ) and the bands were only vestiges of a former structure. Some dehydriding studies on unirradiated SNAP 8 fuel are now being performed to resolve this issue. ^(IV-2) Interpretation of x-ray diffraction examinations has indicated the phases present in dehydrided fuel are those given in Table IV-2. Starting composition had been in the range of 1.72 to 1.78 H/Zr, as used in SNAP 8 endurance tests. Further dehydriding was performed subsequent to the tests, in the course of the present investigation.

The H/Zr values shown in Table IV-2 are subject to verification by chemical analysis, which is not expected to make large changes (i. e., changes greater than ~ 0.1 H/Zr).

On the basis of these results, the α phase is not expected to be present significantly above an H/Zr value of ~ 1.19 .

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TABLE IV-2
PHASES PRESENT IN DEHYDRIDED FUEL

H/Zr _(eff)	α	γ	δ	ϵ	Modifier
1.64	None	Weak	Strong	Difficult to detect	Weak
1.54	None	Weak	Strong	None	Medium
1.44	None	Minor	Strong	None	Weak
1.19	Weak	Moderately strong	Strong	"Strange lines"	None
0.87	Medium	Strong	Strong	None	Very weak

2. Criterion for β -Phase Operation

Any segment of the fuel with a post-test hydrogen analysis of $\langle \text{ZrH}_{1.33} \rangle$, or having a microstructure characteristic of decomposed eutectoid, must have been operating partially in the β phase at time of shutdown. The criterion of a composition $\langle \text{ZrH}_{1.33} \rangle$ comes from the phase diagram (Figure II-2), and the criterion of decomposed eutectoid in the microstructure is self evident. Data for all segments of the fuel meeting either of these criteria are tabulated in Table IV-3.

TABLE IV-3
SEGMENTS OF S8ER FUEL SHOWING EVIDENCE OF β -PHASE OPERATION

Fuel Element	Distance From Top of Fuel Rod (in.)	H/Zr		Micro-structure	$\frac{(\Delta V)_{bu}}{V_i}$ (%)
		Pre-Irradiation	Post-Irradiation		
309	4-1/2	1.69	1.45	$\delta + \gamma$ rim, with $\alpha + \delta$ core	5.81/6.31
332	1-1/2	1.73	0.98	$\alpha + \delta$	-
	7-1/2	1.73	1.35	$\alpha + \delta$	-0.11/0.71
410	12	1.72	1.30	$\delta + \gamma$ rim, with $\alpha + \delta$ core	-

3. Areas of Uncertainty

It is impossible to say with certainty what phases were present during operation in those segments of the fuel having a H/Zr ratio of 1.33 to 1.54, and a microstructure characteristic of $(\delta + \gamma)$ in the postirradiation examination. Phase identification depends on the temperature of operation of that particular segment, and on the degree of rehydriding during shutdown. There is enough uncertainty in both of these variables to make a definite identification impossible. Data on fuel segments meeting these criteria are given in Table IV-4. Again, note the reference to ϵ phase in the microstructure. This is probably dehydrided ϵ (i. e., δ), and not really ϵ .

TABLE IV-4
SEGMENTS OF S8ER FUEL WHICH MAY HAVE OPERATED IN THE
 δ OR IN THE $\beta + \delta$ PHASE

Fuel Element	Distance From Top of Fuel Rod (in.)	H/Zr		Micro-structure	$(\Delta V)_{bu}$ V_i (%)
		Pre-Irradiation	Post-Irradiation		
188	1-1/2	1.76	1.43	$\delta + \gamma$	0.75/1.25
	5-1/2	1.76	1.47	$\delta + \gamma$	0.48/0.87
332	10-1/2	1.73	1.52	$\delta + \gamma$	0.02/0.16
398	11	1.70	1.52	$\delta + \epsilon$	-
476	2-1/2	1.79	1.49	$\delta + \gamma$	1.35/1.69
486	6-3/4	1.77	1.50	$\delta + \gamma$	-
519	6-1/2	1.72	1.52	$\delta + \gamma$	4.22/4.41

B. HYDROGEN LOSS

During the postirradiation examination of the S8ER core, several fuel rods were sectioned diametrically at various elevations and analyzed for hydrogen content as a function of axial location. From the shape of the hydrogen profiles thus obtained, the extent of redistribution of the hydrogen from hot (top) to the cold (bottom) end of the fuel elements was determined. By integrating these profile curves, the total hydrogen remaining in a fuel element at end of life was estimated and compared with pre-irradiation analyses to determine the amount

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Figure IV-1. Change in H/Zr Ratio as Function of Axial Location for Intact Elements

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Figure IV-2. Change in H/Zr Ratio as Function of Axial Location for Cracked Elements

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DISTANCE FROM TOP OF FUEL ROD (in)

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of hydrogen lost during reactor operation. It should be noted that adequate determination of the profile over the 14-in. long fuel rod requires a minimum of probably eight sections, and preferably more. Only five of the fuel rods in Table I-1 met this criterion: 111 (cracked), 188 (intact), 232 (intact), 398 (cracked), and 525 (intact).

Figure IV-1 shows the hydrogen profile data for the intact elements, and Figure IV-2 for the cracked elements. The trend of the data is toward redistribution of hydrogen from the top to the bottom end. Calculated fuel rod centerline temperature profiles for two different radial positions in the core are shown in Figure IV-3.^(IV-3) The two cases selected represent the temperature extremes, and similar plots for other radial positions in the core should fall between these two. One might think the region at the peak centerline temperature should have the lowest hydrogen content, rather than the minimum hydrogen content being at the outlet (top) end; however, the observed hydrogen profiles show the axial distribution of hydrogen is more dependent on surface or radially averaged temperature than on centerline temperature.

The factor of only two between the mean hydrogen losses in cracked and intact elements (0.174 and 0.087, respectively) raises questions as to why the cracked elements retained hydrogen so relatively well. It would seem that, once the cladding ruptures, the glass barrier must also rupture, and there would then be little to prevent excessive dehydrogenation of the fuel rod. Excessive dehydrogenation actually occurred in only two of the eight cracked fuel elements examined: 332 and 398. (Of these, only Element 398 was a "typical" cracked element, having a 4-in. long axial cladding split, starting 4 in. down from the top of the fuel rod). The other cracked elements lost about the same amount of hydrogen as the intact elements.

An evaluation^{*} of the hydrogen loss for intact fuel elements and cracked fuel elements was made on the basis of the H/Zr curves in Figures IV-1 and IV-2. These curves were modified from the actual data results by extrapolating at either end, in order to estimate a total integrated $\Delta(H/Zr)$ for each element. The results of averaging these extrapolations tend to show more of a divergence at the top of the fuel rod between the intact and cracked elements. A statistical

*This evaluation was made by D. S. Burgess

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Figure IV-3. Axial Temperature Distribution for Two Radial Core Locations

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treatment of the total $\Delta(H/Zr)$ values for each element results in a mean of -0.087 and sample variance of 0.00288 for the intact elements, compared to a mean of -0.174 and sample variance of 0.016 for the cracked elements.

Examination of the data was made on the possibility that Elements 398 and 332 might be extreme values which are not representative of the population of cracked elements. This is also indicated for Element 188, with respect to the intact elements. A statistical test for extreme values resulted in not being able to reject Elements 398 and 332 from the cracked element population with greater than 80% confidence.* Surprisingly, however, there is greater than 99.5% confidence that Element 188 belongs in a different population from the rest of the examined intact elements.

These results are even more surprising when it is considered that Element 332 had a weld defect from fabrication, and in other ways (such as extent of dehydriding) shows strong indications of being different from the rest of the cracked elements.

Further examination of the distribution of the changes in H/Zr shows that the variance is so large, for the group of cracked elements, that the distribution is more reasonably described by a "log normal" assumption. This is the assumption that the logarithms of the $\Delta(H/Zr)$ values for each element are distributed normally. Figure IV-4 shows the two relative frequency distributions for the cracked and intact elements. The distribution for intact elements excludes Element 188, which reduces the mean $\Delta(H/Zr)$ to -0.068. It can be seen that the intact elements are grouped very closely about the mean $\Delta(H/Zr)$. However, the cracked element distribution is widely dispersed, even though the mean value is not extremely less than the mean for the intact elements. It is interesting to note that 39% of the distribution for cracked elements lies in the same range as 99.9% of the distribution for the intact elements.

A plausible explanation for the hydrogen retention ability of elements with split cladding is that the surface of the fuel rod was oxidized by the dissolved oxygen in the NaK that came in contact with the rod through the cladding crack. The glass barrier on the cladding served to replenish the oxygen in the NaK,

*This does not rule out the possibility that these elements might be rejected on other criteria, from observations made on them during the detailed examination.

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Figure IV-4. Comparison of $\Delta(H/Zr)$ Frequency Distributions for Cracked vs Intact S8ER Fuel Elements

and so the fuel rod could have been effectively immersed in an oxidizing atmosphere. This served to heal over any breaks that may have occurred in the oxide film. During postirradiation metallographic examination, a surface film ("rind"), which may have been this postulated oxide layer, was found to be present on cracked elements, and was only occasionally observed on intact elements.* The postulated influence of the liquid metal's oxygen content on the retention of hydrogen in hydride fuel is supported by measurements at lower temperatures on unfueled zirconium hydride.^(IV-4) In one test, material with $H/Zr = 1.7$ was heated to 600°C in vacuum, pure sodium, and sodium contaminated with oxygen. At 600°C (1112°F), the loss rate for the first two conditions was about 10^6 times greater than for the third.

*It was actually observed only on Element 410, considered an intact element, but one that was sectioned, not declad. (See Table I-2)

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REFERENCES

- IV-1 Data for Tables IV-1, IV-3, and IV-4 are from unpublished Internal Letters: D. D. McAfee/W. E. Krupp to K. J. Miller, April 25, 1966; D. D. McAfee/W. E. Krupp to K. J. Miller, May 2, 1966; and personal communication, J. B. Vetrano with K. J. Miller, Atomics International, Canoga Park, Calif.
- IV-2 Personal communication with D. Atkins, S. K. Lee, and P. S. Olson, Atomics International, Canoga Park, Calif. (May 13, 1966)
- IV-3 Unpublished Internal Letter, P. M. Magee to E. J. Donovan, "Centerline Fuel Temperature Isotherm Maps for Various S8ER and S8DS Operating Conditions," Atomics International, Canoga Park, Calif. (March 15, 1966)
- IV-4 Enclosure to unpublished letter 393 AT, C. Berke (Interatom) to C. Starr (Atomics International), "Expected Behavior of KNX Moderator Elements under Reactor Operating Conditions," (January 21, 1966)

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V. IRRADIATION-INDUCED VOLUME CHANGES IN SNAP FUEL

By

J. B. Vetrano and M. P. Heisler

The volume changes in the S8ER fuel due to irradiation were examined, to see how they compared with predictions from prior and current capsule irradiation data. The sources of the irradiation capsule data examined are given in Table V-1.

TABLE V-1
CONDENSED SUMMARY OF SNAP FUEL IRRADIATION EXPERIMENTS

Experiment	Description	Approximate Temperature (°F)	Approximate Burnup (met. at. %)	References
NAA 53-1	0.2-in. diameter fuel pins	1300-1470	0.52-0.68	NAA-SR-9633 NAA-SR-MEMO-8191 NAA-SR-MEMO-9182
NAA 53-2	0.2-in. diameter fuel pins	1210-1440	0.95-1.03	
NAA 53-3	0.2-in. diameter fuel pins	1410-1515	0.30-0.36	
NAA 82	1.2-in. diameter fuel pins	1500	0.08	NAA-SR-MEMO-11891
NAA 115-1	0.5-in. diameter fuel pins	1300-1600	0.4	—

In addition to the reference cited in the Table, use was also made of a summary unpublished internal letter, "SNAP Irradiation Programs," by E. M. Chandler to Gillies/Crawford, 10-8-65 (Confidential), and of the series of progress letters from the SNAP 8 Project to the AEC on S8ER and NAA 115 Fuel Element Evaluation.^(I-4)

Three other capsule experiments were considered for the comparison, but not used for various reasons: NAA 26 had too low a burnup, NAA 75 was a low-

temperature experiment with a ternary alloy, and NAA 116 was a low-temperature experiment.

The principal factors considered in the comparison were burnup and volume change. No attempt was made to correlate these properties with temperature. The burnup data are mostly calculated; and, in the cases of long elements (i. e., S8ER and NAA 82), there were significant calculated burnup gradients.

The volume change data were corrected, in all cases, for density changes due to hydrogen loss. The method of making this correction is described in Section III of this report. In many cases, the correction for hydrogen loss was quite substantial, indicative of the necessity of making this correction for any meaningful evaluation of the data.

The data used for the calculation of swelling due to irradiation are given in Table V-2. The column headed " $(\Delta\rho)_{bu}/[\rho_f - (\Delta\rho)_h]$ " is the % volume change due to burnup alone, and is mathematically equivalent to $(\Delta V)_{bu}/V_i$. The data in Table V-2 are plotted as swelling vs burnup in Figure V-1.

Superimposed on the scatter of data points in Figure V-1 is the Miller Correlation for 1500°F operation. This correlation (see NAA-SR-9633) was derived principally from the NAA 53-1, -2, and -3 Experiments, and has been used as a measure of S8ER behavior. A re-examination of the data on which the correlation is based indicates much more uncertainty (in the measurements of density, principally) than would be acceptable with present techniques. Also, the density changes used as input data for the numerical correlation are not corrected for hydrogen loss.

There is considerably more scatter in the data in Figure V-1 than is desirable for sound predictions of SNAP fuel swelling. Certainly, one contributing cause to the scatter is the tacit assumption that the swelling of each small volume of fuel is not influenced by the adjacent material. In cases of axial or radial restraint, such as S8ER, this is certainly not the case. In a gross fashion, however, one can see a consistency in the NAA 82, the NAA 115-1, and the S8ER fuel behavior.

A more quantitative treatment of the data is contained in the next section.

TABLE V-2
IRRADIATION-INDUCED VOLUME CHANGES OF SNAP FUEL
(Sheet 1 of 3)

Experiment	Fuel Element	Distance from Top of Fuel Rod (in.)	ρ_i^* (g/cc)	ρ_f (g/cc)	R_i	R_f	$(\Delta\rho)_h$ (g/cc)	$\rho_f - (\Delta\rho)_h$ (g/cc)	$-(\Delta\rho)_{bu}$ (g/cc)	$\frac{-(\Delta\rho)_{bu}}{\rho_f - (\Delta\rho)_h}$ (%)	Burnup [†] (met. at. %)	
S8ER	111	0 - 1-1/2	6.055	6.034	1.75	1.66	0.009	6.025	0.030	0.50	0.06	
		1-1/2 - 3	6.055	6.084	1.75	1.69	0.006	6.078	-0.023			
		4 - 5-1/2	6.055	6.007	1.75	1.66	0.009	5.998	0.067	1.12	0.188	
		5-1/2 - 7	6.055	5.915	1.75	1.66	0.009	5.906	0.149	2.52	0.205	
		7 - 9	6.055	5.992	1.75	1.69	0.006	5.986	0.069	1.15	0.20	
		10 - 11	6.055	5.968	1.75	1.73	0.002	5.966	0.089	1.49	0.165	
		13 - 13-1/2	6.055	5.991	1.75	1.82	-0.007	5.998	0.057	0.95	0.064	
	188	0 - 1	6.057	6.110	1.76	—	—	—	—	—	0.05	
		1 - 2	6.057	6.083	1.76	1.43	0.101/0.071	5.982/6.012	0.075/0.045	0.75/1.25	0.10	
		2 - 5	6.057	6.073	1.76	1.42	0.106/0.074	5.967/5.999	0.090/0.058	0.97/1.51	0.160	
		5 - 6	6.057	6.086	1.76	1.47	0.081/0.058	6.005/6.028	0.052/0.029	0.48/0.87	0.191	
		6 - 7	6.057	5.962	1.76	1.49	0.070/0.050	5.892/5.912	0.165/0.145	2.45/2.80	0.196	
		7 - 8	6.057	5.938	1.76	1.52	0.054/0.059	5.884/5.899	0.173/0.158	2.68/2.94	0.193	
		8 - 9-1/2	6.057	5.962	1.76	—	—	—	—	—	—	
		9-1/2 - 10-1/2	6.057	5.993	1.76	1.62	0.014	5.979	0.078	1.30	0.165	
		10-1/2 - 14	6.057	5.995	1.76	1.73	0.003	5.992	0.065	1.09	0.105	
		206	0 - 3-1/2	6.093	6.071	1.75	1.62	0.013	6.058	0.035	0.58	0.103
			10-1/2 - 11-1/2	6.093	6.015	1.75	1.72	0.003	6.012	0.081	1.35	0.145
	232	1 - 2	6.092	6.044	1.73	1.53	0.048/0.034	5.996/6.010	0.096/0.082	1.36/1.60	0.105	
		2 - 3-1/2	6.092	5.991	1.73	1.52	0.052/0.038	5.939/5.953	0.153/0.139	2.34/2.58	0.155	
		3-1/2 - 5	6.092	5.940	1.73	1.60	0.013	5.927	0.165	2.79	0.188	
		5 - 6	6.092	5.900	1.73	1.56	0.032/0.024	5.868/5.876	0.224/0.216	3.68/3.82	0.205	
		6 - 8-1/2	6.092	6.018	1.73	1.63	0.010	6.008	0.084	1.40	0.214	
		10-1/2 - 11-1/2	6.092	6.000	1.73	1.72	0.001	5.999	0.093	1.55	0.152	
		12 - 14	6.092	6.030	1.73	1.81	-0.008	6.038	0.054	0.90	0.085	

*Nomenclature — See Nomenclature, Section III.

†Burnup values from postirradiation examinations by: S8ER - K. J. Miller, 115-1 — R. E. Forrester, 82-1 — T. A. Park

TABLE V-2
IRRADIATION-INDUCED VOLUME CHANGES OF SNAP FUEL
(Sheet 2 of 3)

Experiment	Fuel Element	Distance from Top of Fuel Rod (in.)	ρ_i^* (g/cc)	ρ_f (g/cc)	R_i	R_f	$(\Delta\rho)_h$ (g/cc)	$\rho_f - (\Delta\rho)_h$ (g/cc)	$-(\Delta\rho)_{bu}$ (g/cc)	$\frac{-(\Delta\rho)_{bu}}{\rho_f - (\Delta\rho)_h}$ (%)	Burnup [†] (met. at. %)
S8ER	293	5 - 7	6.09	6.03	1.74	1.63	0.011	6.02	0.07	1.15	0.200
		12 - 14	6.09	6.027	1.74	1.79	-0.005	6.032	0.06	1.0	0.076
	309	1 - 2	6.094	6.058	1.69	1.39	0.116/0.079	5.942/5.979	0.152/0.115	1.92/2.56	0.115
		3 - 5	6.094	5.817	1.69	1.45	0.085/0.058	5.732/5.759	0.362/0.335	5.81/6.31	0.195
		10 - 14	6.094	6.007	1.69	1.64	0.005	6.002	0.092	1.53	0.125
	332	1 - 2	6.093	6.470	1.73	0.92					
		6 - 8-1/2	6.093	6.203	1.73	1.33	0.153/0.103	6.050/6.100	0.043/0.007	-0.11/0.71	0.20
		9-1/2 - 14	6.093	6.123	1.73	1.54	0.040/0.031	6.083/6.092	0.010/0.001	0.02/0.16	0.12
	342	4 - 7	6.093	5.902	1.72	1.53	0.047/0.033	5.855/5.869	0.238/0.224	3.81/4.06	0.237
	378	2-1/2 - 3-1/2	6.089	5.823	1.69	1.58	0.011	5.812	0.277	4.76	0.150
	398	6 - 8	6.089	5.872	1.70	1.52	0.048/0.025	5.824/5.847	0.265/0.242	4.14/4.55	0.234
		12 - 14	6.089	6.056	1.70	1.59	0.011	6.045	0.044	0.73	0.090
	432	3 - 5	6.094	5.863	1.74	1.61	0.013	5.850	0.244	4.17	0.168
		9 - 10	6.094	6.033	1.74	1.67	0.007	6.026	0.068	1.13	0.17
		10-1/2 - 12	6.094	6.034	1.74	1.76	-0.002	6.036	0.058	0.96	0.130
	474	3 - 4	6.106	5.947	1.80	1.70	0.010	5.937	0.169	2.84	0.205
		7 - 9	6.106	5.972	1.80	1.70	0.010	5.962	0.144	2.41	0.236
		11 - 12	6.106	5.984	1.80	1.77	0.003	5.981	0.125	2.09	0.16
	476	1-1/2 - 3-1/2	6.091	6.063	1.79	1.49	0.073/0.053	5.990/6.010	0.101/0.081	1.35/1.69	0.14
		5-1/2 - 7-1/2	6.091	6.057	1.79	1.60	0.019	6.038	0.053	0.88	0.205
		8-1/2 - 9-1/2	6.091	6.004	1.79	1.63	0.016	5.988	0.103	1.72	0.19
	519	0 - 2	6.094	6.021	1.72	1.44	0.093/0.065	5.928/5.956	0.166/0.138	2.32/2.40	0.09
		5-1/2 - 6-1/2	6.094	5.873	1.72	1.55	0.037/0.026	5.836/5.847	0.258/0.247	4.22/4.41	0.228
		11 - 13	6.094	6.030	1.72	1.71	0.001	6.029	0.065	1.08	0.14

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TABLE V-2
IRRADIATION-INDUCED VOLUME CHANGES OF SNAP FUEL
(Sheet 3 of 3)

Experiment	Fuel Element	Distance from Top of Fuel Rod (in.)	ρ_i^* (g/cc)	ρ_f (g/cc)	R_i	R_f	$(\Delta\rho)_h$ (g/cc)	$\rho_f - (\Delta\rho)_h$ (g/cc)	$-(\Delta\rho)_{bu}$ (g/cc)	$-(\Delta\rho)_{bu}$ $\rho_f - (\Delta\rho)_h$ (%)	Burnup [†] (met. at. %)
S8ER	525	1 - 2	6.086	6.069	1.69	1.55	0.031/0.023	6.038/6.046	0.048/0.040	0.66/0.80	0.11
		2-1/2 - 3-1/2	6.086	6.028	1.69	1.58	0.011	6.017	0.069	1.15	0.155
		3-1/2 - 4-1/2	6.086	5.991	1.69	1.60	0.009	5.982	0.104	1.74	0.180
		4-1/2 - 6	6.086	5.998	1.69	1.62	0.007	5.991	0.095	1.59	0.20
		8 - 9	6.086	5.950	1.69	1.63	0.006	5.944	0.142	2.39	0.195
		10-1/2 - 11	6.086	5.997	1.69	1.58	0.011	5.986	0.100	1.67	0.155
		11 - 12	6.086	5.969	1.69	1.77	-0.008	5.977	0.109	1.82	0.135
		12-1/2 - 14	6.086	6.009	1.69	1.82	-0.013	6.022	0.064	1.06	0.080
115-1	1A	Center	6.050	—	1.68	1.47	0.050-0.073	—	—	—	0.39
	2	Center	6.099	5.9796	1.59	1.49	0.035-0.053	5.945-5.927	0.154-0.172	2.59-2.90	0.41
	3A	Center	6.061	5.8554	1.66	1.61	0.005	5.850	0.211	3.61	0.41
	4	Center	6.100	5.8559	1.71	1.65	0.006	5.850	0.250	4.28	0.41
	5A	Center	6.061	5.8271	1.67	1.63	0.001	5.826	0.235	4.04	0.41
	6	Center	6.071	5.9766	1.69	1.53	0.030-0.042	5.947-5.935	0.124-0.136	2.08-2.29	0.43
	7	Center	6.053	5.8799	1.84	1.51	0.052-0.067	5.828-5.813	0.225-0.240	3.86-4.13	0.41
	8	Center	6.062	6.0170- 6.0054	1.82	1.45	0.070-0.097	5.947-5.908	0.115-0.154	1.93-2.60	0.42
	9	Center	6.068	5.9679- 5.9648	1.68	1.62	0.006	5.962-5.959	0.106-0.109	1.78-1.83	0.43
	10	Center	6.073	5.9843	1.69	1.61	0.008	5.976	0.097	1.62	0.42
82-1	11	Center	6.073	5.9665	1.67	1.64	0.003	5.964	0.110	1.84	0.39
	12	Center	6.072	5.9763	1.69	1.66	0.003	5.973	0.009	1.66	0.37
		0.8	6.065	6.005	1.84	1.90	-0.006	6.011	-0.054	0.90	—
		2.7	6.065	6.027	1.84	1.86	-0.002	6.029	-0.036	0.60	0.07
		4.4	6.065	6.003	1.84	1.77	0.007	5.996	-0.069	1.15	0.14
		6.5	6.065	5.956	1.84	1.71	0.013	5.943	-0.122	2.06	0.14
		8.0	6.065	5.986	1.84	1.71	0.013	5.973	-0.092	1.54	0.06
		10.1	6.065	6.005	1.84	1.71	0.013	5.992	-0.073	1.22	0.06
		11.5	6.065	6.037	1.84	1.81	0.003	6.034	-0.031	0.51	—

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Figure V-1. Distribution of Swelling in SNAP Fuels (Miller Correlation Prediction for 1500°F shown for comparison)

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VI. ANALYSIS OF IRRADIATION-INDUCED VOLUME CHANGE DATA

By
C. K. Smith

A. INTRODUCTION

Any attempt to devise a reliable empirical correlation for irradiation-induced volume change in SNAP fuel must take into account the following conditions:

- 1) The large number of variables possibly involved (burnup, temperature, carbon content, H/Zr ratio, U content, burnup rate, mechanical constraints, etc.)
- 2) The lack of statistically reliable data from controlled irradiation experiments over the range of variables possibly involved, as discussed elsewhere in this report
- 3) The unavailability of measured temperature and burnup data, which requires the use of calculated temperature and burnup curves
- 4) The necessity for analytical separation of hydrogen loss or gain effects from burnup effects, with the assumption that hydrogen effects are completely reversible and separable.

Several past attempts^(VI-1, -2, -3) to devise empirical correlations to describe SNAP fuel behavior have proven to be unsuccessful in predicting the volume changes observed in the S8ER fuel. Despite these difficulties, the importance of being able to make reasonably accurate predictions of fuel behavior makes it also important that existing correlations be re-examined whenever significant new data become available. This section presents the beginnings of such a re-examination, the conclusions reached, and recommendations for further work.

The NAA 115-1 and S8ER data shown in this section are those tabulated in Section V. The NAA 82-1 points were obtained by plotting the raw data,^(VI-4) making hydrogen corrections as appropriate, using the techniques presented in Section III. From the resulting curves, the data points were extracted at 10% length increments between 20 and 70% of the 82-1 fuel rod length, measured from the top of the rod.

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B. GENERAL DISCUSSION

A graph of observed $(\Delta V)_{bu}/V_i$ as a function of burnup for NAA 82-1, NAA 115-1, and intact S8ER elements is shown in Figure VI-1. If one examines that graph, together with the data associated with each point, several tentative and qualitative observations can be made:

- 1) Fuel growth appears to be a strong function of burnup; and, at low burnups, reasonably independent of temperature.
- 2) Fuel growth appears to be a function of carbon content; this is most evident in the 115-1 results.
- 3) Fuel growth appears to be a weaker function of temperature than of burnup and carbon content, over the ranges of those variables included in the data; this is most evident in comparison of swelling observed at the top and bottom of each S8ER rod.
- 4) Fuel growth might be a function of $H/Zr_{(eff)}$; but such a dependence will be difficult to establish, because of the manner in which $H/Zr_{(eff)}$ varies within each fuel rod during operation.
- 5) Dependence of fuel growth on uranium content and burnup rate is indeterminate, from the available data, because of the small range of these variables in the data.
- 6) The influence of mechanical restraint (by adjacent fuel and, in some cases, the element cladding) is not easily assessable from the data; however, examination of the data from 115-1 indicates the volume increase of a radially restrained short fuel rod (IS-6 had zero final radial clearance) is of the same order as the volume increases of short fuel rods having no external restraint.

On the basis of these observations, it appears the general form selected for a first attempt at correlating the 82-1, 115-1, and S8ER burnup swelling can at least contain terms to account specifically for burnup, composition, and temperature effects.

As presented in Section VII, the fuel volume change attributable to burnup has several components. These are considered individually, in the following, to arrive at a model suggested for further work toward correlation of burnup volume change.

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One would expect that, for a perfect lattice or one which is sufficiently homogeneous, the fuel volume change attributable to fission product volumes might be directly proportional to the burnup [i. e., $(\Delta V)_{fp \text{ vol}}/V_i = Ab$]. However, the S8ER fuel contains lattice imperfections resulting from several factors, including the dispersion nature of the fuel and the presence of carbon, which could possibly cause the coefficient and/or exponent of b in the lattice equation to be functions of composition. For example, preliminary examination of the NAA 116-1 and -2 low-temperature irradiation data^(VI-5) suggests such a compositional dependence might exist. Therefore, it appears the model for the correlating equation should include this term as

$$(\Delta V)_{fp \text{ vol}}/V_i = f(c) b^{n(c)} , \quad \dots(VI-1)$$

where f(c) and n(c) are functions of fuel composition to be determined.

Swelling attributable to fission gas agglomeration might be dependent upon burnup, temperature, fuel composition, and other parameters. Data over a significant range of values are available only for the three parameters just mentioned specifically, and all other parameters will thus show up in an empirical correlation as lumped constants which cannot be separated until sufficient data is available. The fission gas agglomeration term for fixed fuel composition would then consist of a constant, times a factor to account for gas availability, times a fractor to account for agglomeration rate [i. e., for a volume of fixed composition, a term such as $B b^m \exp(-T_a/T)$ appears representative of fission gas agglomeration]. Of the "constants" in that term, it appears the constant (B) and the activation temperature (T_a) have some probability of a compositional dependence, while (m) does not. Therefore, it appears the model for the correlating equation should contain the fission gas agglomeration term as

$$(\Delta V)_{fg \text{ agglom}}/V_i = g(c) b^m \exp[-h(c)/\bar{T}] , \quad \dots(VI-2)$$

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where $g(c)$ and $h(c)$ are possible functions of fuel composition and are to be determined. Use of an average fuel temperature implies the fuel volume being examined is selected in such a way that temperature variations within the volume are small.

Unfortunately, the knowledge of underlying phenomena is too incomplete to allow even qualitative formulation of a term to account for cavitation swelling or micro-tearing (see Section VII). However, it is also not yet clear that cavitation swelling contributed appreciably to the volume changes observed in SNAP fuel.

C. SUGGESTED FORM OF CORRELATING EQUATION

Combining Equations VI-1 and -2, the form suggested for further work to correlate SNAP fuel data is

$$(\Delta V)_{bu}/V_i = f(c)b^{n(c)} + g(c)b^m \exp[-h(c)/\bar{T}] \quad \dots(VI-3)$$

In cases where the volume being considered cannot be selected such that temperature variations within the volume are small, the quantity

$$\frac{1}{\text{volume}} \int_{\text{volume}} \exp[-h(c)/T] d(\text{volume})$$

should be used in place of $\exp[-h(c)/\bar{T}]$ in Equation VI-3.

Trial values suggested for initiation of further work are $f(c) = 6$ to 7 , $n(c) = 0.75$ to 1 , $m = 2$, and $h(c) = 20,000$ to $30,000$, for b in met. at. % and T in °R. These values are based on a very preliminary examination of the S8ER and NAA 115-1, 82-1, and 116-1 and -2 irradiation volume change data.

D. SAMPLE CALCULATION OF REQUIRED AXIAL AND DIAMETRICAL GAPS

The following example illustrates the use of burnup volume change estimates, such as could be produced by a correlating equation, to size the diametrical and axial gaps required in a hypothetical fuel element containing a fuel rod

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0.5 in. in diameter, 14.5 in. in length, and having temperature and burnup distributions as illustrated in Figure VI-2.

Figure VI-2. Temperature and Burnup Profiles Used in Example Calculations

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Dividing the rod length into 10 equal increments, the initial length of rod in each increment is $L_{i,j} = 1.45$ in., the initial diameter of rod in each increment is $D_{i,j} = 0.5$ in., and the initial volume of rod in each increment is $V_{i,j} = 1.45 \pi / 16$ in.³. From Figure VI-2, the average temperature and burnup in each increment is as follows:

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<u>z/L</u>	<u>$\bar{T}$ (°R)</u>	<u>b (met. at. %)</u>
0.0 - 0.1	1800	0.160
0.1 - 0.2	1830	0.220
0.2 - 0.3	1845	0.265
0.3 - 0.4	1850	0.305
0.4 - 0.5	1835	0.325
0.5 - 0.6	1805	0.325
0.6 - 0.7	1765	0.305
0.7 - 0.8	1725	0.265
0.8 - 0.9	1690	0.220
0.9 - 1.0	1655	0.160

Assume the volume increase attributable to burnup in each increment is as follows:

<u>z/L</u>	<u>$\left[(\Delta V)_{bu} / V_i \right]_{\text{design}, j}$ (%)</u>
0.0 - 0.1	1.35
0.1 - 0.2	1.91
0.2 - 0.3	2.40
0.3 - 0.4	2.87
0.4 - 0.5	3.00
0.5 - 0.6	2.79
0.6 - 0.7	2.48
0.7 - 0.8	1.99
0.8 - 0.9	1.62
0.9 - 1.0	1.21

The design values used should include factors necessary to account for the statistics on the correlating equation.

Applying Equation III-12 with these volume growths,

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$$\left[\frac{(\Delta D)_{bu}}{D_i} \right]_{\text{design},j} \leq 1.00\%$$

in all increments. Therefore, for the temperature and burnup conditions of this example, a diametrical gap of 5 mils is adequate to accommodate diametrical growth of the hypothetical 0.5-in. diameter rod. From Equation III-13,

$$\left[\frac{(\Delta L)_{bu}}{L_i} \right]_{\text{design},j} \leq \frac{1}{2} \left[\frac{(\Delta V)_{bu}}{V_i} \right]_{\text{design},j}$$

in each interval. Multiplying both sides of the latter equation by $L_{i,j}$ and summing over the rod length gives

$$[(\Delta L)_{bu}]_{\text{design}} = \sum_{j=1}^{10} [(\Delta L)_{bu}]_{\text{design},j} \leq \sum_{j=1}^{10} \frac{L_{i,j}}{2} \left[\frac{(\Delta V)_{bu}}{V_i} \right]_{\text{design},j}$$

which, for the conditions of this example and the tabulated volume growths, indicates a required axial gap of 0.16 in. to accommodate axial growth.

The diametrical and axial gaps calculated, using this technique, are those which give a high probability that the cladding will not be strained at all, as a result of fuel burnup growth during the fuel element lifetime. If the initial diametrical gap in the fuel element of the preceding illustration were made <5 mils, as a result of either manufacturing tolerances or intentional reduction, the maximum hoop strain which would occur in the cladding as a result of burnup swelling would be 0.2% per mil reduction in diametrical gap, which corresponds to 0.01% strain per 1% reduction in diametrical gap.

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NOMENCLATURE

<u>Symbol</u>	<u>Quantity</u>
A, B, m	Constants
b	Burnup
D	Diameter
f(c), g(c), h(c), n(c)	Functions of fuel composition
L	Length
T	Temperature
T _a	Activation temperature
$\bar{T}$	Average temperature
V	Volume
z/L	Fractional distance from top of fuel rod
<u>Subscripts</u>	
bu	Attributable to burnup
design	Design value; includes factors necessary to account for statistics on correlating equation
fg agglom	Attributable to fission gas agglomeration
fp vol	Attributable to fission product volume
i	Initial
j	Value in j th increment

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VII. IRRADIATION PHENOMENA IN S8ER FUEL

By
W. V. Johnston

A. SUMMARY OF FABRICATION PROCESS^(VII-1)

S8ER fuel is made from triple consumable-arc melted, double-extruded zirconium alloy rod, containing about 10 wt % of 93% enriched uranium and a few tenths wt % carbon (added in the form of ZrC).^{*} Hydrogen is added at 900°C by a gaseous diffusion process to a minimum of 1.7 wt % of total alloy. The massively hydrided product is cut and ground to nominal final dimensions of 0.532 in. diameter and 14.00 in. length.

As the ratio of hydrogen atoms to zirconium atoms (H/Zr) grows to its final value of over 1.7 during the hydriding process, uranium is rejected from the alloy, and the zirconium is converted to the face-centered tetragonal, ϵ -phase hydride.

The ZrC is essentially insoluble in zirconium hydride, and acts as a nucleating agent to control grain size.^(VII-2, -3) Without it, large columnar grains of the ϵ phase are formed, with their long axes oriented along the fuel rod radius, resulting in a low yield of uncracked fuel rods. The additive also improves the tensile strength of the fuel, the maximum increase coming at about 0.15 wt % C.^(VII-4) The composition of the additive in the finished fuel is a carbohydride of the form ZrC-ZrH_x , where $x < 2$; but the additive particles appear to have a core of carbide whose composition is $\text{ZrC}_{0.54}$.^(VII-5) Although exact stoichiometry has not yet been worked out, it is clear that the combination of zirconium with carbon reduces the number of zirconium atoms available for combination with hydrogen. The number of "effective" zirconium atoms is assumed to be the total in the alloy, minus the number of carbon atoms. "Effective" ratios, $\text{H/Zr}_{(\text{eff})}$, (see footnote, Table I-1) are computed from the effective numbers of zirconium atoms, and are used throughout this report without the subscript.^(VII-4, -6)

^{*}Some of the S8ER fuel rods had graphite added, but ZrC is presently the preferred form of additive.

B. AS-FABRICATED MICROSTRUCTURE^(VII-2)

At room temperature, the uranium appears as a dispersion of small, α -phase particles, essentially randomly distributed in the hydride matrix within grains and on grain boundaries. The average uranium particle size is about 0.5μ . Their density is about 5×10^{10} particles/cc. The additive particles are usually found within hydride grains, and vary in average size from $\sim 5 \mu$ at 0.2 wt % C composition to 14μ at 0.5 wt % C. Their density is of the order of 10^8 particles/cc. The hydride is face-centered tetragonal, with a c/a ratio of ~ 0.927 . The c/a ratio varies with hydrogen content, becoming progressively smaller than unity as H/Zr increases beyond ~ 1.5 .^(VII-6)

C. GENERAL CONSIDERATIONS ON IRRADIATION EFFECTS

S8ER fuel reached peak burnups near 0.2%, expressed as percentage of heavy metal atoms. Table VII-1 contains a listing of atom concentrations in nominal composition fuel, and shows that this burnup corresponds to fissioning of 4.8% of the uranium atoms, or about 1 in 20.

TABLE VII-1
ATOM CONCENTRATIONS IN NOMINAL S8ER FUEL COMPOSITION
[Starting Composition (wt %): 89.8 Zr - 10U - 0.2C]

	Numbers of Gram Atoms	Fraction of Total Atoms	Atom %
Zr	0.985	0.943	94.3
U	0.0425	0.0407	4.07
C	0.0167	0.0159	1.59
Total	1.044	1.00	100
Metal atoms	1.028		
0.2% of metal atoms	0.002056 $\approx$ 4.8% of U atoms		

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When any two-phase fuel is under irradiation, there is always the possibility that the structure will become unstable, and will tend toward single-phase under the action of thermal or displacement spikes. This tendency is proportional to the fission rate, temperature, and solubility of the precipitate in the major phase.

The average fission rates in S8ER fuel at reactor powers of 600 and 400 kw were 1.7×10^{12} and 1.1×10^{12} fissions/cc-sec, respectively. It is concluded that the solubility of carbon or uranium in zirconium hydride is low enough so that, at the fission rates indicated, no substantial re-solution of the precipitates is likely to occur.

The average range of fission fragments is 6.8μ in uranium and 9.1μ in zirconium. (VII-7) It is probably greater than this in ZrH_x . The uranium particle size and spacing in S8ER fuel, and the fission fragment ranges, are such that all fission fragments will be expected to recoil out of the originating particle, and into other particles and the adjacent hydride matrix. The distribution of fission recoils, and thus the fission damage, will be homogeneous. In the subsequent discussion, the S8ER fuel will be treated as though it were a homogeneous alloy fuel, with two important exceptions. First, the lattice changes that underlie the observed dimensional changes are those of the zirconium hydride structure, not the dispersed uranium fuel. Second, because of the variation in H/Zr in a specified volume arising from permeation loss and/or hydrogen redistribution, structure changes occur that are related only to compositional change. (See Section III)

The Task Force has concluded that the cracking of the cladding was due to stresses imposed upon it by the fuel, rather than other means such as thermal stresses. We must now consider the mechanisms by which the fuel expanded out against the cladding. The design diametrical clearance between the fuel and the cladding was 3 mils, with an uncertainty of ± 1 mil. Analysis of diametrical increases ($\% \Delta D/D$) in intact elements shows that strains up to 0.9% were tolerated before cracking began. (VII-8) Assuming isotropic growth, the equivalent volume change is about 2.7%. (If the growth in the radial direction is greater than estimated on the assumption of isotropy, the volume change would be less than this.) Supposing that the cladding would have permitted an additional strain of $\sim 0.4\%$ before reaching the point of fracture, the estimated equivalent volume change at fracture would be $\sim 3.9\%$. Many of the S8ER elements showed changes of this magnitude, as did the 115-1 capsules.

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On the scale of fuel swelling phenomena, this is such a small change as to make it difficult to distinguish among the possible causative mechanisms. Evidence of these mechanisms is only likely to become apparent in light-microscope metallographic postirradiation examination of fuel if the volume changes are greater than about 8%. Metallographic examination of the 82-1, 115-1, and S8ER fuel microstructure has thus far, in every case, shown phases present that are directly related to what would be expected for an unirradiated specimen of the same composition. With the exception of portions of S8ER Elements 332 and 398, which had lost large amounts of hydrogen, there is no evidence of cracking or voids in grain boundaries, nor have any bubbles been observed such as are attributable to fission gas.

On the basis of present theories of volume increase in irradiated fuel, there are three causes which must be considered in evaluating S8ER fuel. These are fission product volumes, fission gas atom agglomeration, and cavitation. Superimposed on these are the phenomena associated with phase changes (β - δ and δ - ϵ in both directions) in the zirconium hydride caused by variations in H/Zr and temperature. These causes are discussed separately in the following sections, but all of them may have been operative simultaneously in at least some S8ER elements.

D. DISCUSSION OF SOURCES OF VOLUME CHANGE

1. Fission Product Volumes

These are simply the changes due to new atoms produced in fission, which are larger than the uranium they replace. In S8ER fuel, the rate of volume increase from this source is estimated to be 2.2% per met. at. % burnup. At 0.2 met. at. % burnup, this would produce a volume increase of about 0.45%. Clearly, this cannot account for much of the observed volume increase; but it provides a level to which all other increases should be referred.

2. Fission Gas Agglomeration

The nucleation and growth of fission gas bubbles is usually the major cause of nuclear fuel volume increases, if the fuel operating temperature is high enough to permit fission product diffusion at a significant rate, and if the burnup is high enough. It appears that, at about 1400°F, the S8ER fuel may have been at the

lower boundary of the temperature range for significant diffusion. This conclusion is supported by the relative insensitivity of the observed % ($\Delta V/V$) values in S8ER and 115-1, over the temperature range of 1300 to 1550°F. (VII-9) Thus, some means of bringing gas atoms together may be of more importance in S8ER fuel than thermal diffusion. Examples of such mechanisms are moving dislocations or grain boundaries, fuel thermal stresses, or fuel temperature gradients, all of which are likely to be enhanced by reactor events, such as scrams or power transients that deviate from steady-state operation. The manner in which this may happen has been considered by Barnes and Nelson, (VII-10) and may be summarized as follows:

The fission gases, krypton and xenon, may be assumed to be very insoluble in zirconium hydride. As a consequence, solubilities will be exceeded in the first few minutes of reactor operation. Collisions between gas atoms, or between gas atoms and microstructural features of the hydride, will result in stable bubble nuclei which will grow with the addition of gaseous atoms subsequently produced. Initially, the largest portion of nuclei form on dislocations. This initial nucleation density is extremely high, of the order of 10^{16} nuclei/cc. Simple calculations indicate that, if this density of nuclei (and therefore bubbles) can be maintained, large amounts of these gases can be contained in them at high pressures, and only minor (<1%) volume changes can occur. If, however, the bubbles are brought together by some other means, such as dislocation or grain boundary motion (caused by the previously mentioned sudden stress or temperature changes), they will join together, forming a single larger bubble at a lower gas pressure. Table VII-2 shows quantitatively what happens as the number of bubbles is reduced but the amount of gas is held constant. It is also evident, from the table, that the average bubble radius — say, 2000 Å — results in a bubble size that is below the resolving power of a light microscope at 500X. This indicates why porosity due to bubble formation could not be observed in the examination of S8ER fuel in a light microscope at 500X. Electron microscopy must be employed.

3. Phase Changes

a. $\delta \rightarrow \beta$ Phase Change in Zirconium Hydride

As hydrogen is removed from δ -phase zirconium hydride to below an H/Zr ~ 1.5 , the body-centered cubic β phase is nucleated in the existing face-centered-

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TABLE VII-2
VOLUME CHANGES ASSOCIATED WITH FISSION GASES

Equivalent Burnup (met. at. %)	No. of Fission Gas Atoms/cc	No. of Bubbles/cc	Radius of Bubbles (Å)	$\Delta V/V$
0.1	10^{19}	10^{14}	130	0.12
0.1	10^{19}	10^{13}	400	0.25
0.1	10^{19}	10^{12}	1200	1.25
0.5	5×10^{19}	10^{15}	100	0.5
0.5	5×10^{19}	10^{14}	300	1.25
0.5	5×10^{19}	10^{13}	800	2.5
0.5	5×10^{19}	10^{12}	3000	12.5

cubic δ phase. As the volume of the new phase increases at the expense of the δ phase, the interface between them must move through the solid. In the presence of a temperature gradient, the β phase may move up the temperature gradient and tend to concentrate in one area. This sweeping of a boundary will gather together along the boundary all gas bubbles which are not attached to precipitate particles. The bubbles may join together by diffusing along the boundary. This phenomenon may be the overriding cause of fuel volume increases in any fuel in which it occurs. Examples where this has happened are the U-Nb and U-Mo^(VII-11) and U-Zr^(VII-12) systems.

Willis and Moses^(VII-13) have shown that there is a sudden increase in the release of fission gases from zirconium hydride fuel as the δ phase transforms to the β phase. Their samples, 0.004 in. thick and 0.25 in. in diameter, had a much greater surface-to-volume ratio than the S8ER fuel rods. The enhanced release therefore only suggests a mechanism which could cause volume swelling in other geometries. They found that the effect of phase change upon the release of fission gas was relatively greater, the higher the burnup prior to the phase change. Such a rate effect suggests a relatively greater swelling if the phase

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change occurs later in the irradiation (i.e., the swelling would increase with burnup at a greater than linear rate). Their burnups ranged from about 10^{15} to 10^{17} fissions/cc, which correspond to relatively short periods of operation of the S8ER. At burnup levels from 1 to 6×10^{15} fissions/cc, they found that material with 0.11% carbon released less gas than 0.0 or 0.40% material. This supports the possibility that carbon content affects swelling (See Section VI). Finally, they observed that the $\delta \rightarrow (\beta + \delta)$ transformation direction caused more gas release than the reverse.

Fission gas release measurements on S8ER fuel showed that only a small quantity was released. Based on Kr^{85} , only ~0.015% of that produced was detected when the cladding was punctured. This is less than the amount calculated from surface recoil.^(VII-14) It is possible that gas and other fission products were trapped in the glass barrier. We suggest that annealing experiments on the glass might provide additional information about the fission gas release from the fuel.

S8ER Element 398 showed metallographic evidence of β phase, in the upper few inches of the fuel rod, consistent with hydrogen composition analysis. Porosity and crack formation are also visible in this region. This porosity is not necessarily related to gas effects, since the β phase is more dense than the δ phase, and transformation may cause grain boundary tearing and pulling away. There were only a few S8ER elements which had lost sufficient hydrogen to show metallographic evidence of β phase. Thus, while it is possible that a $\delta \rightarrow \beta$ phase transformation may have caused cracking in a few rods, there were a number of cracked rods which should never have approached the δ - β phase boundary and some uncracked rods which lost much hydrogen. (See Section IV).

Postirradiation annealing experiments, combined with electron microscopy and with dehydriding/rehydriding experiments on unirradiated fuel, can clarify the relationship between fission gas behavior, phase change, and microstructure.

b. $\epsilon \rightarrow \delta$ Phase Change in Zirconium Hydride

Zirconium dihydride has a face-centered-tetragonal crystal structure with c/a ratio < 1 . As this ϵ phase loses hydrogen, there is a transformation to the face-centered-cubic δ phase. There is considerable uncertainty in the exact nature of the transformation, and in particular, the temperature dependence of it.

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(See Section II). Most workers in the field consider the transformation to be a shear type, rather than a first-order transformation, as is a nucleation and growth type of transformation.^(VII-13) This type of transformation, while it involves a large change in c/a ratio (nearly 10%), does not have the ability to sweep gas atoms along with it. Willis and Moses^(VII-13) did not find any gas release on passing through this transformation. This is fortunate, because nearly all of the S8ER fuel underwent this transformation, in one direction or the other, in some portion of the rod during the irradiation. Repeated cycling through this transformation may be a source of fuel instability, because each grain undergoes a dimensional change on transformation; and repeated cycling may result in a ratcheting effect, such as is found in thermal-cycled uranium. This effect would be expected to be more pronounced in coarse-grained hydride not containing the carbon additive.

It is not possible to make further comments about this transformation, because of a lack of information. It is one of the recommendations of this review that additional research be pursued on the nature and range of this transformation and the effect of thermal cycling through it. A density increase of about 1% has been reported on transforming from δ to ϵ .^(VII-15)

4. Cavitation Swelling

The third major source of volume increases in nuclear fuels is an interaction between the gas bubbles or gas atoms, and "growth" (change of shape without change in density) that takes place in massive uranium metal.*

In massive α -phase uranium, growth arises from anisotropy of the orthorhombic structure, which has different coefficients of thermal expansion along the three crystallographic axes. It occurs at temperatures below $\sim 950^\circ\text{F}$ (i.e., below the temperature at which self diffusion is sufficiently rapid to anneal the mixture of vacancies and interstitials generated by the passage of fission fragments through the lattice). In the proper fuel temperature range, stresses due to anisotropic thermal expansion within the fission spikes force the vacancies and interstitials to aggregate on different crystal planes. During irradiation

*It is emphasized that it is the zirconium hydride that may behave like massive uranium metal. The dispersed uranium phase present in SNAP fuel is not involved.

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below 950°F, a uranium crystal expands in the $\langle 010 \rangle$ direction and contracts in the $\langle 100 \rangle$ direction. The dimensional change in a practical fuel shape, such as a rod, is greater for larger grain, more highly oriented material. (VII-16)

Growth is manifested at relatively low burnups, because fission products are not involved in the mechanism.

It has been shown that anisotropic growth during irradiation by fission fragments occurs with other metals having noncubic structure, such as Zr, Zn, Cd, and Ti. (VII-17, -18, -19) The growth is ascribed to differential thermal stresses, similar to those occurring in uranium, arising from the structural anisotropy.

Because ϵ -phase zirconium hydride has a tetragonal crystal structure, the possibility of growth exists in S8ER fuel. The coefficients of expansion along the c and a axes are not known, but it is expected that they would not be equal. Such expansion measurements are preferably made on single crystals. Accurate knowledge of the coefficients would help elucidate phenomena in ϵ -phase fuel.

Interestingly, fast neutron irradiation alone does not induce dimensional or compositional changes in unfueled, ϵ -phase zirconium hydride. No significant change was observed in H/Zr = 1.7 material following exposure to about 10^{21} neutrons/cm² (energy >1 Mev), over the temperature range from 70 to 1050°F. (VII-20) Fast neutron irradiation also does not result in dimensional changes in the noncubic metals, where Buckley (VII-19) has shown that direct exposure to fission fragment bombardment (as in contact with fissioning fuel) is needed to produce the effect. It would be worthwhile to do experiments on the structural changes in unfueled zirconium hydride, similar to Buckley's. A fission fragment source similar to that described by Engberg (VII-21) may be suitable for such tests.

Growth in ϵ -phase hydride fuel, as in other media, would result in a change in shape but no volume (density) change. If, in a polycrystalline fuel rod, there is an appreciable preferred orientation of grains in either axial or radial directions, the crystalline anisotropy would act to close any gap to the cladding more rapidly than would be expected, on the assumptions of random grain orientation and isotropic swelling (see Sections III and VI).

At higher burnup, where there is an appreciable inventory of fission gas, and at temperatures above the range where growth along predominates, interaction

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occurs between the fission gas and the anisotropic dimensional change. This may be called cavitation swelling, or micro-tearing. It has been observed in uranium at temperatures between 1050 and 1100°F, where recovery and recrystallization begin;^(VII-22) and is also similar in mechanism to that of void formation during high temperature creep tests.^(VII-23, -24, -25) This model, in its crudest form, suggests that, on irregularities (precipitates, gas bubbles, etc.) in grain boundaries, sufficiently high tensile stresses are generated during grain boundary sliding to nucleate small voids. These voids then grow by vacancy and gas atom condensation. While boundary sliding and cavitation in a conventional creep test are caused by an externally applied load, the driving force for cavitation swelling is the internal stress within and between adjacent grains, caused by the crystallographic anisotropy. The net result of this process is an increase in volume, which may include a shape change (depending upon the degree of anisotropy) residual from the growth phenomenon.

The cavitation mechanism of volume increases is of interest to the S8ER evaluation. The fuel operating temperature is in the right range. Although the degree of anisotropy of the starting ϵ phase would probably result in only a small effect, and even though the fuel dehydrided later to the cubic δ structure, the voids nucleated during the early portion of the irradiation would serve as locations that would collect gas and grow more rapidly than other fission gas bubbles. These voids would also be so small as to be invisible at 500X; but, if examined with an electron microscope, there should be sufficient difference in their alignment and orientation, compared to the fission gas bubbles, that an evaluation of the two mechanisms might be made.^(VII-26, -27)

E. BASIS FOR FUEL SWELLING CORRELATION

At present, it is not possible to determine the dominant mechanism for S8ER fuel volume increases. Adding to the fission product volume increase, both fission gas bubbles and cavitation voids may be operative, so that there may not be a single dominant mechanism. The effects of net changes in H/Zr that result in no phase change, or in a phase change, must be separately considered. Therefore, it does not appear feasible to develop, from considerations of underlying phenomena, quantitative expressions to relate volume change with composition, burnup, and temperature. The data are best handled empirically (See Section VI).

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(There was no Page 72 in original document.)

APPENDIX

INTERNAL LETTER

ATOMICS INTERNATIONAL
*(Division of North American Aviation, Inc.)*TO . Those Listed
ADDRESS .
*(Show Dept., Group,
Div. & Location)*

DATE . February 10, 1966

FROM . C. Starr
ADDRESS . 797 Canoga-2

PHONE . 2201

SUBJECT . S8ER Fuel Evaluation Task Force

The SNAP 8 Experimental Reactor Program was extremely successful from the point of view of surpassing peak temperature and endurance objectives; however, the extended reactor operation resulted in gross cracking of the fuel element cladding. It is essential that the behavior of the 8ER fuel be sufficiently understood to preclude the possibility of a similar occurrence in the SNAP 8DS reactor. Therefore, a full understanding of the 8ER fuel behavior is a high priority task with major long-range program implications. The critical importance of this task dictates the full utilization of all technical skills available within AI.

Therefore, a technical task force headed by Dr. H. Pearlman is hereby established to augment the substantial efforts currently applied to this problem. Specifically, the task force is established for the purpose of:

- a) insuring that the hot cell post-operational examination of 8ER fuel elements is continually pointed in the direction of acquiring the most significant available data,
- b) summarizing and evaluating all currently available information on hydride material and its irradiation stability in order to provide a sound basis for evaluating the 8ER behavior,
- c) insuring that the reactor nuclear, thermal, and hydraulic characteristics have been sufficiently defined to identify the range of operating conditions experienced by the fuel,
- d) insuring that all available data are subjected to detailed scrutiny and cross correlation with the significant fuel material properties and the reactor operating parameters in an attempt to deduce the cause of the fuel behavior,
- e) identifying and providing technical data and analysis to support the probable cause or causes of the detailed 8ER fuel behavior,
- f) postulating those experiments or proof tests which can add critical additional information to the understanding of the 8ER behavior, provide the basis for eliminating hypothesis, and demonstrate the validity of the final analysis of the problem.

IL To: Those Listed
From: C. Starr
Subject: S8ER Fuel Evaluation Task Force

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February 10, 1966

The following personnel are assigned to work under the direction of Dr. Pearlman towards the above objectives:

D. Burgess
M. P. Heisler
C. K. Smith
J. B. Vetrano
W. V. Johnston (NAA Science Center)

The purpose of the task force is to provide technical leadership and coordinated judgment to the final resolution of the S8ER problem. The current personnel assignments and responsibilities of the functional organization are purposefully preserved. Close cooperation and support from the continuing efforts of the functional organization are mandatory for success. The task force will report to D. Mason, SNAP 8 Project Manager, in order to insure complete coordination with the SNAP 8 program and to take advantage of the authority of the Project Manager's office.

The task force is targeting for the culmination of this effort into a final report on April 1, 1966. Your complete cooperation and support is required for the successful completion of this important effort.

C. Starr
C. Starr