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NASA TM X-2224

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(NASA-TM-X-2224) CALCULATION OF RADIATION INDUCED SWELLING OF URANIUM MONONITRIDE USING THE DIGITAL COMPUTER PROGRAM CYGRO 2
H.W. Davison, et al (NASA) Mar. 1971 15 p
CSCL 18J H1/22
N72-33652
Unclas
43889

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CALCULATION OF RADIATION INDUCED SWELLING OF URANIUM MONONITRIDE USING THE DIGITAL COMPUTER PROGRAM CYGRO-2

by Harry W. Davison and Ivan B. Fiero

Lewis Research Center

Cleveland, Ohio 44135

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • MARCH 1971

[redacted]

15P.

[redacted]

[redacted]

1. Report No.
NASA TM X-2224

2. Government Accession No.

3. Recipient's Catalog No.

4. Title and Subtitle
CALCULATION OF RADIATION INDUCED SWELLING OF URANIUM MONONITRIDE USING THE DIGITAL COMPUTER PROGRAM CYGRO-2

5. Report Date
March 1971

6. Performing Organization Code

7. Author(s)
Harry W. Davison and Ivan B. Fiero

8. Performing Organization Report No.
E-5951

9. Performing Organization Name and Address
Lewis Research Center
National Aeronautics and Space Administration
Cleveland, Ohio 44135

10. Work Unit No.
120-27

11. Contract or Grant No.

12. Sponsoring Agency Name and Address
National Aeronautics and Space Administration
Washington, D. C. 20546

13. Type of Report and Period Covered
Technical Memorandum

14. Sponsoring Agency Code

15. Supplementary Notes

16. Abstract
Fuel volume swelling and clad diametral creep strains were calculated for five fuel pins, clad with either T-111 (Ta-8W-2.4Hf) or PWC-11 (Cb-1Zr-0.1C). The fuel pins were irradiated to burnups between 2.7 and 4.6 atomic percent. Clad temperatures were between 1750° and 2400° F (1228 and 1589 K). The maximum percentage difference between calculated and experimentally measured values of volumetric fuel swelling is 60 percent.

~~NOFORN~~

17. Key Words (Suggested by Author(s))
Uranium nitride
Fuel swelling
Nuclear fuels

18. Distribution Statement
Confidential - limited

19. Security Classif. (of this report)
Confidential

20. Security Classif. (of this page)
Unclassified

21. No. of Pages
15

22. Price

~~[redacted]UP 1 Exc[redacted] from au[redacted]atic downgra[redacted] declassification~~

[redacted]

[redacted]

[redacted]

CALCULATION OF RADIATION INDUCED SWELLING OF URANIUM MONONITRIDE USING THE DIGITAL COMPUTER PROGRAM CYGRO-2

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SUMMARY

A lithium-cooled, uranium mononitride (UN) fueled reactor is being considered as a thermal power source for a space electric power system. This reactor is designed to operate for 50 000 hours with fuel burnups between 2.5 and 4 percent and at fuel clad temperatures between 1700° and 2200° F (1200 and 1478 K). Under these conditions volumetric fuel swelling and clad diametral creep strains must be determined to design fuel pins for the reactor. The CYGRO-2 digital computer program was used to calculate fuel volume swelling and clad diametral creep strain in five UN fuel pins which had been irradiated at the Oak Ridge National Laboratory. These fuel pins were clad with either T-111 (Ta-8W-2.4Hf) or PWC-11 (Cb-1Zr-0.1C) and operated at burnups between 2.7 and 4.6 percent and clad temperatures between 1750° and 2400° F (1228 and 1589 K). Of the fuel pins irradiated at ORNL, these five pins most closely resembled those in the lithium cooled UN fueled reactor.

Assuming that the UN surface tension is 1500 dynes per centimeter and the fission gas bubble density is 1×10^{15} bubbles per cubic inch (0.6×10^{14} bubbles/cm^3), the calculated volumetric fuel swelling and clad creep agreed closely with measured values. The maximum percentage difference between measured and calculated values is 60 percent.

Although these results are encouraging, only five experiments have been investigated and there is uncertainty in many of the parameters used in the calculations such as fuel surface tension, fuel and clad mechanical properties, and component temperatures.

INTRODUCTION

The NASA Lewis Research Center is investigating a uranium mononitride (UN) fueled fast reactor (ref. 1) which operates with fuel clad temperatures of between 1700° and 2200° F (1200 and 1478 K) and fuel burnups between 2.5 and 4 percent. This reactor is

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cooled with liquid lithium and is designed as a thermal power source for a space electric power system. A primary problem area in this reactor which is also common to many power reactors is fuel element failure due to excessive fuel swelling. It is desirable to develop and utilize a mathematical model which will predict volumetric fuel swelling and diametral clad swelling in order to design fuel pins with a low probability of failure. It would be an advantage to base this model on experimental data. If swelling of the experimental fuel pins can be predicted, then the model can be used to predict swelling in proposed fuel pin designs with more confidence. Little irradiated fuel swelling data in this temperature range is available in the literature except for the irradiation tests conducted at the Oak Ridge National Laboratory under the SNAP-50 program (refs. 2 and 3).

The purpose of this report is to describe the analytical and experimental UN fuel volume swelling and diametral clad creep strains and compare the calculated results with the experimental results. Experiments selected for comparison were those most closely resembling the Lewis mononitride fueled fast reactor operating conditions.

Several investigators have developed fuel swelling and fission gas migration models. Barnes (ref. 4) and Greenwood and Speight (ref. 5), for example, developed models to estimate the unrestrained swelling in ceramic fuels based on bubble migration and coalescence. Friedrich and Guilinger (ref. 6) developed a digital computer program (CYGRO-2) which neglects the effects of bubble migration and coalescence but includes the effect of the clad material in suppressing fuel swelling. Since the fuel swelling of the fuel pins of interest is greatly dependent on clad restraint, the CYGRO-2 computer program was utilized to calculate fuel pin swelling.

The reactor experiments are described first, followed by a brief description of the swelling model. The calculational approach is then presented and the results of the comparisons between calculations and experiments are given. Values of the parameters used in the CYGRO-2 program are presented in the appendix.

DISCUSSION OF REACTOR EXPERIMENTS

Uranium mononitride (UN) fuel irradiations were conducted by the Oak Ridge National Laboratory (ORNL) in the Low Intensity Test Reactor (LITR) and the Materials Test Reactor (MTR). The fuel pin designs and irradiation test results from the LITR and MTR are summarized in references 2 and 3, respectively. The fuel pin samples were 1/4-inch- (0.63-cm-) diameter right circular solid cylinders. The fuel was physically separated from the 0.025-inch- (0.063-cm-) thick clad by a 0.005-inch- (0.013-cm-) thick layer of tungsten, which was chemically vapor deposited on the inside surface of the clad. These experimental samples consisted of T-111 (Ta-8W-2.4Hf) and PWC-11 (Cb-1Zr-0.1C) clad fuel pins. The fuel pins were irradiated to burnups between 2.7 and 4.6 atomic percent and at clad temperatures between 1750° and 2400° F (1228 and 1589 K). Total irradiation time was between 7000 and 12 000 hours. The operating parameters and measured values of fuel volume change and clad diametral creep strains are summarized in table I. Fuel volume changes represent measured averages for each pin but maximum for the capsule and range between 5.6 and 11 percent. The clad diametral creep ($\Delta D/D$) values are the maximum changes measured in any fuel pin capsule. Clad creep strain values of up to 1.7 percent were measured.

DESCRIPTION OF MODEL

The fuel volumetric swelling and clad creep strains in the irradiated fuel specimens were calculated using the CYGRO-2 digital computer program. This program was written by Friedrich and Guilinger of Bettis Atomic Power Laboratory (ref. 6) and was modified by Fiero of NASA Lewis Research Center (ref. 7). The CYGRO-2 program allows calculation of the time-dependent stresses and strains in clad cylindrical fuel pins operating at either steady-state or transient power conditions. The program includes elastic, plastic, and creep analyses to calculate the effects of various loading conditions.

Fuel swelling is assumed to be caused by the expansion of stagnant fission product gas bubbles and the formation of solid fission products. Fuel swelling is retarded by the creep strength of the fuel, the surface tension around the fission gas bubbles, and the restraining force provided by the clad after the fuel contacts the clad. Other parameters included in the analyses are thermal gradients in the fuel and clad, pressure outside the clad, and the pressure between the fuel and clad. The sources of the parameters used in the calculations are given in the appendix.

RESULTS

The CYGRO-2 program was used to calculate fuel volume swelling and clad diametral creep in the five irradiated pins shown in table I. Since some of the parameters used in the CYGRO-2 program are not well known, a special calculational approach was taken. The calculations were made by (1) initially using best estimates of these parameters to calculate swelling for the five fuel pins, (2) adjusting the parameters for a particular reference pin (LN-3B) to determine which caused the greatest changes in fuel swelling and clad creep, and (3) the most critical parameter was adjusted (within its known range) to improve the agreement between calculations and measurements. All five pins were then recalculated with this adjusted parameter.

The four parameters investigated are the following:

1. Fuel surface tension

2. Fission gas bubble density (bubbles/in.^{3} (bubbles/cm^3) of fuel)
3. Amount of fission gas leaking out of the fuel
4. Increase in creep strength of the clad due to radiation hardening

In attempting to compare calculations with experimental data it has been found that there are additional uncertainties not related to these parameters. The uncertainties exist for both the experimental and calculated data. First, a given experimental capsule usually contains more than one pin irradiated under the same conditions; consequently, there exists a range of fuel and clad swelling measurements. (Table I only shows the maximum measured values.) Secondly, there is an uncertainty in the size of the fuel-clad gap. Therefore, the approach in the comparison was to compare ranges, where the range in experimental data was compared to a range of calculated data based on the smallest and largest possible gap sizes.

The results of the initial calculations of fuel swelling ($\Delta V/V$) and clad creep strains ($\Delta D/D$) for the five experiments shown in table I are compared in figure 1(a) with measured values. The 45° line indicates perfect agreement. Experiment numbers are shown both in table I and figure 1(a). The rectangles shown in the figure represent uncertainties in both calculated and measured values. The measured uncertainties (vertical side of rectangle) represent differences in measurements on pins irradiated within the same capsule. In those cases where no vertical lines are shown (experiment LN-3B) there was only one fuel pin in the capsule. The calculated uncertainties (horizontal lines) of figure 1(a) are due only to the uncertainty in the fuel-clad gap. Due to fabrication tolerances there was a 0.001-inch (0.003-cm) uncertainty in the size of the gap. Note, however, that there are uncertainties in the calculations in addition to the fuel-clad gap which are not illustrated in figure 1(a). These uncertainties are represented by the input parameters to CYGRO-2 (such as the four previously listed) which are not well known, but which are discussed later.

The fuel swelling results tend to be conservative (calculated values higher than measured values) in experiments LN-3B (T-111 clad), 57-665, and 57-003 (PWC-11 clad). These experiments experienced either clad temperatures above 2000° F (1366 K) or burnups above 4 percent. The values of clad creep tended to be slightly conservative only in experiments LN-3B and 57-003. Both of these experiments experienced clad temperatures above 2000° F (1366 K).

The four previously listed input parameters used in the CYGRO-2 program were changed to determine their effect on the calculated values of UN volumetric swelling and clad creep strain. Experiment LN-3B having the T-111 clad and the highest temperature was selected as a reference case for the comparison. The UN volumetric swelling and T-111 clad diametral creep strain initially calculated with 0.001-inch (0.003-cm) gap were 14.4 and 2.2 percent, respectively. The corresponding measured values are 8.9

and 0.8 percent. The effects of each of the variables are summarized in table II and discussed subsequently.

Surface Tension

The surface tension was increased by a factor of three (from 500 to 1500 dynes/cm). This increase in surface tension reduced the fuel swelling by 35 percent and reduced the clad creep strain by 41 percent. Although the value of the surface tension of UN is not known, surface tension values for uranium or uranium compounds between 500 and 1500 dynes per centimeter have been measured. The former value was measured for UO_2 (ref. 8); the latter value was measured for U (ref. 9).

Bubble Density

The number of fission gas bubbles per cubic inch of fuel was increased from 1×10^{15} to 27×10^{15} (0.6×10^{14} to 16×10^{14} bubbles/cm^3). This change produces the same reduction in fuel swelling and clad creep as was found when the surface tension was increased by a factor of 3. This is expected because the Laplace stress σ_b caused by surface tension is

$$\sigma_b = \frac{2\gamma}{r}$$

The total bubble volume is

$$V_T = N_H \frac{4}{3} \pi r^3$$

$$\sigma_b \propto \gamma N_H^{1/3}$$

where

γ surface tension

r radius of one fission gas bubble

N_H number of fission gas bubbles per unit volume

V_T total volume of fission gas bubbles

Therefore, a factor of 3 increase in surface tension would be expected to cause the same change in fuel swelling and resulting clad creep strain as a factor of 27 increase in bubble density.

Gas Leakage

The gas leakage from the fuel pellets was doubled (increased from 5 to 10 percent), resulting in only 4-percent reduction in fuel swelling and 4-percent reduction in clad creep. The amount of gas leakage from the fuel pins in the five experiments was measured and was generally less than 10 percent.

Radiation Hardened Clad

Clad creep strength, tensile strength, and brittleness generally increase during exposure in a radiation environment. Radiation hardening effect in T-111 clad material has not been measured. In order to obtain an indication of the importance of this effect, it was assumed that during the irradiation period the creep strength doubled, varying linearly with burnup from its unirradiated strength. This creep strength is a measure of the creep rate at constant stress; that is, the strength is considered doubled if the creep rate is halved at the same stress. The tensile strength was not changed.

Doubling the creep strength of the clad reduced the final fuel swelling by only 4 percent. The total clad diametral creep strain was unaffected.

Fuel-Clad Gap

The gap between the fuel and clad was increased from 0.001 to 0.002 inch (0.003 to 0.005 cm). Increasing the fuel-clad gap had a negligible effect on the fuel volumetric swelling but the clad creep strain is reduced 32 percent. Because the fuel temperature is known from measurements, it was not allowed to change when the fuel-clad gap was increased.

The previous comparisons indicate that fuel volumetric swelling and clad creep strain are most sensitive to fuel surface tension and bubble density. The fuel pin swelling was recalculated for the five experiments shown in table I assuming a fuel surface tension of 1500 dynes per centimeter. The measured and calculated values of fuel volumetric swelling and clad creep strain are compared in figure 1(b). Increasing the surface tension of UN from 500 to 1500 dynes per centimeter improved the agreement between calculated and measured values of fuel swelling and clad diametral creep strain. The maximum disagreement between calculations and measurements is noted in experiment 57-003 where the calculated volumetric fuel swelling (16.5 percent) is 60 percent greater than the measured fuel swelling (10.2 percent). Although only five experiments have been calculated and many of the variables are not well known, the agreement between calculations and measurements is acceptable.

CONCLUSIONS

The radiation induced swelling of 96-percent dense UN fuel and diametral creep strain of T-111 and PWC-11 clad were calculated using the CYGRO-2 digital computer program. These calculated values were compared with the experimental values to determine the applicability of the CYGRO-2 program for use as a design and analysis tool. Agreement between analytic and experimental fuel swelling is considered good, especially since experimental conditions and material properties were not always well defined.

Only four of the input properties and/or conditions were considered uncertain enough to warrant a series of runs to determine their effect. These were fuel surface tension, gas bubble density, fission gas release, and the irradiation strengthening of the clad creep strength. It was found that only surface tension and gas bubble density (both of which affect gas bubble equilibrium) has any significant effect on the fuel volumetric swelling. Consequently, the surface tension was increased which improved agreement between calculated and experimental fuel swelling.

Since there was usually more than one fuel pin in a capsule, a range of swelling data exists for supposedly identical fuel pins. One parameter which could have varied amongst the pins within a given capsule is the fuel-clad gap. The calculations were done with both maximum and minimum possible gap sizes, and this range compared with the experimental data.

It was also concluded from the limited number of experiments checked that the greater the burnup the more likely that CYGRO-2 is to predict greater than experimentally measured swelling. This makes the program conservative from a design standpoint.

Assuming a fission gas bubble density of 1×10^{15} bubbles per cubic inch (0.6×10^{14} bubbles/cm^3) and a fuel surface tension of 1500 dynes per centimeter, the maximum percentage difference between calculated and measured fuel swelling values is 60 percent.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, October 20, 1970,
120-27.

APPENDIX - DISCUSSION OF PARAMETERS USED IN CALCULATIONS

Numerous parameters are required as input to the CYGRO-2 program for the fuel swelling and clad creep calculations. These parameters can be categorized as fission gas production parameters, physical property parameters, and mechanical property parameters.

Fission Gas Production Parameters

The fission gas concentration within the fuel is calculated in the CYGRO-2 program using a simple rate equation:

$$\frac{dM}{dt} = A\dot{\varphi} - BM$$

where

M lb moles of gas/in.^{3} of fuel

A lb moles of gas/fission

$\dot{\varphi}$ fissions/hr/in.^{3} of fuel

B gas leakage coefficient, hr^{-1}

t time, hr

The first term, dM/dt, represents the rate of change of gas concentration in the fuel. The second term represents the total gas production rate due to fissions. The third term represents the rate of gas leakage out of the fuel material.

The value of A was calculated assuming that 0.3 molecule of fission gas is produced per fission (ref. 10) and that 82 percent (ref. 11) of the nitrogen in the UN recombines within the fuel. Therefore, 0.39 molecule of gas is produced per fission. Although this value is somewhat temperature dependent, it was assumed constant in all of the calculations. The value of A, therefore, is 1.426×10^{-27} moles per fission. The value of B was calculated from the measured values of gas leakage from the experiments. The gas leakage values were generally between 5 and 10 percent. Gas pressure in the fission gas bubbles was calculated assuming a bubble density in the fuel of 1×10^{15} bubbles per cubic inch (0.6×10^{14} bubbles/cm^3).

Observable bubble densities in a variety of fuel compositions (pure and impure uranium and UO_2) varies from as low as 5.5×10^{13} bubbles per cubic centimeter to as high as 10^{17} bubbles per cubic centimeter (refs. 12 to 14). ORNL observed that most of the bubbles remained within the fuel grains and not at grain boundaries. Therefore, only one type of bubble was assumed in this analysis. In addition, the number of bubbles assumed result in bubble diameters comparable to those observed by ORNL within the irradiated fuel.

Contribution of solid fission products to fuel swelling can be estimated on a theoretical basis. This was done (ref. 15) for UO_2 with a nominal growth rate of 0.35-percent $\Delta V/V$ for 10^{20} fission per cubic centimeter (0.864 percent $\Delta V/V$ per percent burnup). The solid fission product growth was assumed to be 0.85 percent $\Delta V/V$ per percent burnup for these calculations.

Physical Property Parameters

References for the physical property parameters are summarized in table III. Although surface tension of UN has not been measured, values of 500 and 1500 dynes per centimeter were assumed. The fission gas properties in the bubbles contained in the fuel were calculated using the van der Waals equation of state assuming the fission product gas is xenon. The van der Waals constants for xenon were obtained from reference 16.

Mechanical Property Parameters

References for the mechanical properties are summarized in table III. The fuel and clad creep phenomena are calculated in CYGRO-2 using

$$\dot{\epsilon} = 10^{C}\left(\frac{\sigma}{1000}\right)^{G}$$

The plastic stress-strain relation is calculated from

$$\epsilon = 10^{P}\left(\frac{\sigma}{1000}\right)^{Q}$$

where

σ stress, psi (N/cm^2)

ϵ strain

$\dot{\epsilon}$ strain rate, hr^{-1}

The C, G, P, and Q are empirical parameters which are fitted to the creep and stress-strain data.

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TABLE I. - OAK RIDGE NATIONAL LABORATORY FUEL SWELLING EXPERIMENTS

Irradiated pins	Clad material[a]	Time, hr	Burnup, percent	Temperature, $^{o}F(K)$		Fuel swelling,[b] percent	Clad diametral creep,[b] percent
				Clad	Fuel		
LN-3T	T-111	7 045	2.8	1815(1264)	2000(1366)	5.6	<0.4
LN-3B	T-111	7 045	2.8	2380(1578)	2570(1683)	8.9	.8
57-665	PWC-11	9 533	4.6	1770(1239)	2100(1422)	8.0	1.2
57-669	PWC-11	10 352	2.7	1795(1253)	1965(1347)	7.0	.4
57-003	PWC-11	11 985	4.2	2160(1455)	2415(1597)	11.0	1.7

[a]Clad composition: T-111, Ta-8W-2.4Hf; PWC-11, Cb-1Zb-0.1C.
[b]Maximum values measured.

TABLE II. - EFFECT OF VARIOUS CYGRO INPUT PARAMETERS ON FUEL SWELLING

Parameter	Fuel swelling, $\Delta V/V$, percent	Change in fuel swelling, percent	Clad diametral creep, $\Delta D/D$, percent	Change in diametral creep, percent
Reference experiment: LN-3B[a]	14.4	0	2.2	0
3$\overline{X}$ Surface tension	9.3	-35	1.3	-41
27$\overline{X}$ Bubble density	9.3	-35	1.3	-41
2$\overline{X}$ Gas leakage	13.8	-4	2.1	-4
Radiation hardened clad (1/2 ϵ)	13.8	-4	2.2	0
2$\overline{X}$ Fuel-clad gap	14.3	-1	1.5	-32

[a]0.001-inch (0.003 cm) radial fuel-clad gap.

TABLE III. - SOURCES OF MECHANICAL AND

PHYSICAL PROPERTY DATA

Physical property parameters	Data sources		
	UN	T-111	PWC-11
Coefficient of thermal expansion	17	21	25
Elastic modulus	17	21	21
Poissons ratio	17		
Stress-strain properties			26
Creep properties	17, 18, 19	22, 23, 24	
Thermal conductivity	20	21	21

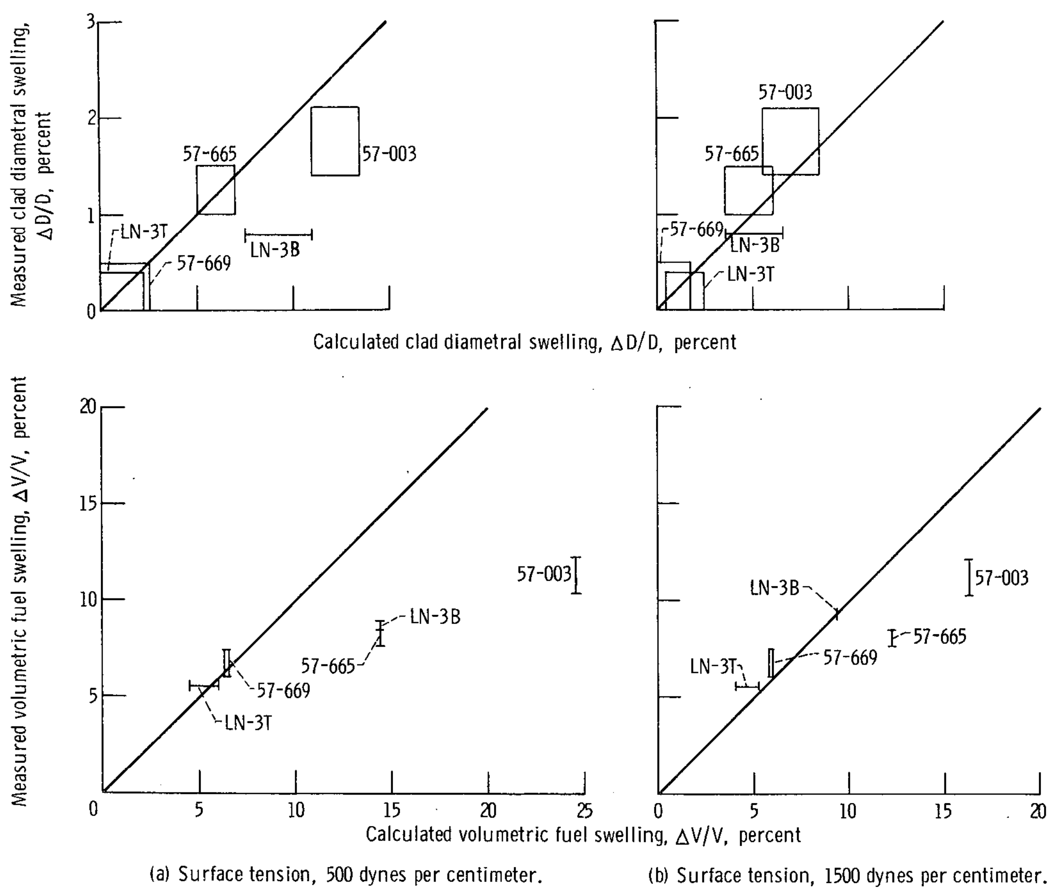

Figure 1. - Comparison of measured and calculated fuel swelling (CYGRO). Calculated uncertainties due to fuel-clad gap size; measured uncertainties due to pin variation per capsule.

NASA-Langley, 1971 — 22 E-5951