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STATUS REPORT I ON THE TESTING OF GRAPHITE-TYPE [NUCLEAR [ROCKET (ROVER)] FUEL IN A PULSED REACTOR (TREAT)

(Title Confidential Restricted Data)

by

Robert C. Liimatainen, Richard O. Ivins,
Max A. Vogel and Frank J. Testa

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DATE AUG 20 1973

For The Atomic Energy Commission

Bram C. Feldman

Bram C. Feldman

Chief, Reactor, Space and Technology Branch

Division of Classification RESTRICTED DATA

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Nuclear Reactors for
Rocket Propulsion
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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
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STATUS REPORT I ON THE TESTING OF GRAPHITE-TYPE
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Robert C. Liimatainen, Richard O. Ivins,
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Chemical Engineering Division

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Nuclear Reactors for Rocket Propulsion

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I. INTRODUCTION

The behavior of the core material during reactor transients is of particular interest in the field of nuclear rockets. At the startup of the rocket engine, it is important to know how rapidly the power level can be increased (as measured by the period or rise time) while still retaining the integrity of the fuel. Under other circumstances, for example re-entry, it is desirable to achieve destruction of the reactor core into small particles. One method of accomplishing this "destruct," which is under consideration, is to cause the core to undergo such an energetic reactor pulse that it effectively disintegrates the fuel into small particles which can be burned up on atmospheric re-entry.

To obtain some information on the fuel which is under development for the Rover project (land-based tests of reactor cores for this project are known as the KIWI series; the final flight engine will be known as Nerva), a co-operative program was started with the following participants: Argonne National Laboratory (ANL), Los Alamos Scientific Laboratory (LASL), and the Westinghouse Astronuclear Laboratory (WANL). The role of these 3 organizations in the work to be reported was roughly as follows:

- ANL: furnished experimental facilities, manpower, and consultation and coordination of the experimental work.
- LASL: furnished fuel specimens, guidance, and outline of program and radiochemical analyses.
- WANL: furnished design and procurement assistance together with personnel who helped in all phases of the work. Provided program coordination so as to integrate the experimental work with the objectives of Nerva flight safety.

Thus far in the TREAT program of the ANL Chemical Engineering Division, metals, cermets, and ceramic fuels (oxides) have been studied;

all of these are of interest in water-cooled nuclear reactors.¹ The program was, therefore, expanded to include a type of fuel (graphite or carbide) which is of primary interest in high-temperature, gas-cooled reactors (the KIWI and Nerva engines use hydrogen as the coolant and thrust producing gas).

II. EXPERIMENTAL

Six experimental assemblies containing an enriched-uranium-bearing graphite fuel were subjected to transient nuclear reactor bursts in TREAT² at the National Reactor Testing Station in Idaho. Following the experiments, the in-pile assemblies were returned to Argonne, Illinois, for postirradiation examination. The fuel specimens had the following characteristics:

weight:	4.0 gm
dimensions:	cylinders of 0.5-in. diameter x 0.75-in. length with 7 longitudinal coolant holes, 0.1 in. in diameter
density:	2.1 gm/cc
U ²³⁵ loading:	0.4 gm U ²³⁵ /cc
type of fuel:	shelf (also referred to as "air stored" or "hydrolyzed" since some of the uranium in the graphite is present as UO ₂); analyses by LASL show that these shelf-type fuel specimens had about 25 percent of the uranium as the oxide (UO ₂) and 75 percent as the carbide (UC ₂).

The fuel specimen was contained within a stainless steel autoclave in a 20-psia helium atmosphere, thus providing a closed system with an inert environment. The fuel specimens were located inside graphite crucibles and were radially supported by small guides. The fuel material was, however, free to expand during the transient.

Figure 1 shows a diagram of the internal assembly of these experiments and also a photograph of the autoclave and the various components. The autoclave was, in turn, contained within a Zircaloy-2, dummy TREAT fuel can. Graphite reflector blocks were located both above and below the

¹R. C. Liimatainen, R. O. Ivins, M. F. Deerwester, and F. J. Testa. Studies of Metal-Water Reactions at High Temperatures II. TREAT Experiments: Status Report on Results with Aluminum, Stainless Steel-304, Uranium, and Zircaloy-2, ANL-6250 (1961).

²F. Kirn, J. Boland, H. Lawroski, and R. Cook, Reactor Physics Measurements in TREAT, ANL-6173 (1960).

autoclave (inside of the fuel can), and also a split graphite cylinder was placed around the body of the autoclave; the autoclave was, therefore, completely surrounded by graphite.

Figure 1

AUTOCLAVE FOR IN-PILE TESTS OF GRAPHITE FUEL SPECIMENS

INTERNAL ASSEMBLY

VIEW OF COMPONENTS

III. RESULTS AND DISCUSSION

Table 1 gives a summary of the data from the first 2 series of experiments with graphite fuel specimens arranged in order of increasing energy of the reactor burst. The data indicate that there is a threshold of destruction between 250 and 350 Mw-sec, which corresponds to a peak fuel temperature between 1700°C and 2000°C. This means that, for those transients in which the maximum temperature of the fuel is less than 1700°C, the fuel will remain intact, whereas more energetic transients will produce fragmentation of the graphite fuel.

Table 1

SUMMARY OF EXPERIMENTS CONDUCTED IN TREAT ON SHELF-TYPE
GRAPHITE FUEL SPECIMENS IN A HELIUM ENVIRONMENT

CEN Transient No.	Reactor Pulse			Appearance of Fuel Pin after Transient	Mean Particle Size, d_{sv} , mils	Results		Peak Temperatures of Fuel Pin, °C
	Peak Power, Mw	Integrated Power, Mw-sec	Period, msec			Xe133 Released from Fuel ^b		
						μ C	%	
79	302	120	139	intact; no change	500	6	1	850
80	696	250	93	some cracking at top; cut retained basic cylindrical shape	500	34	3	(1700) ^C
73	2390	several transients ^a	150 to 54	destruction into frag- ments and particles	62	4700	44	2500-final transient
74	2420	390	54	destruction into frag- ments and particles	32	1400	73	>1500 (2150) ^C
75	3820	650	40	destruction into frag- ments and particles	23	2100	66	>1500 (2900) ^C
76	3850	890	40	destruction into frag- ments and particles	22	2300	51	no thermocouple (3350) ^C

^aA plot of observed temperature versus reactor energy for CEN-73, shows a slight "knee" in the curve at about 2.0 to 2.5 energy units, thus suggesting breakup of the fuel in that range (see Figure 4).

^bMeasured from gas sample of autoclave and corrected back to time of irradiation.

^cValue obtained from calibration experiment (CEN-73) of temperature versus energy input (see Figure 4). The total energy of the reactor pulse is correlated to the energy absorbed by the fuel pin by the relationship $\frac{2.6 \text{ fissions}}{(\mu\text{g U235})(\text{watt-sec})}$ or equivalently, $\frac{3.5 \text{ cal/gram of pin}}{\text{Mw-sec}}$. This factor is based on burnup determinations performed by LASL personnel on the irradiated specimens from CEN-74, 75, and 76.

Figure 2
TRANSIENT CEN-79
GRAPHITE FUEL
SPECIMEN

1/2 in.
Side view

Transient CEN-79 was a relatively low-energy reactor pulse (120 Mw-sec); the fuel pin (see Figure 2) remained completely intact while attaining a peak temperature of 850°C. There were no observable changes in appearance of the fuel pin, such as cracking or distortion. Transient CEN-80 was a 250-Mw-sec burst on a 93-msec reactor period; this pulse produced a maximum fuel pin temperature of 1700°C. Observation of the fuel pin after the transient showed that the specimen was intact; however, some cracking was apparent and a small fragment had fallen off from the top of the pin. Figure 3 shows both end and side views of the fuel pin after CEN-80. It cannot be unequivocally stated that the observed cracking was caused by the heating and cooling during the reactor pulse. There is a chance that some mechanical force (such as from the spring pushing on contact points on the fuel) may have induced the cracks. Further work will be done to bracket more closely the temperature (or corresponding energy input) which causes destruction of these air-stored graphite fuel specimens.

Figure 3
TRANSIENT CEN-80
GRAPHITE FUEL SPECIMEN

Several transients were conducted with the fuel pin in CEN-73. A tungsten-7 percent rhenium, tungsten-26 percent rhenium thermocouple was located in the fuel pin to measure its temperature. A calibration of the energy input from the reactor was accomplished by increasing successively the integrated power of the transients; these results are shown in Figure 4.

Figure 4
A COMPARISON OF OBSERVED AND CALCULATED TEMPERATURES
FOR A GRAPHITE FUEL SPECIMEN AT
VARIOUS REACTOR ENERGIES (CEN-73)

Figure 4 also gives a comparison of the calculated adiabatic temperatures with the actual, measured temperature rise of the fuel. The data in Figure 4 indicate that the fuel specimen is heated (starting from ambient room temperature at the initiation of the transient) in nearly an adiabatic manner up to temperatures of about 1500°C. At the higher temperatures, the measured maximum temperature attained by the test specimen becomes progressively less than the calculated adiabatic temperature. Thus, there is a suggestion of significant heat loss, possibly by radiation, at the higher energies; another factor which might have caused the indicated temperature to be lower is a partial loss of contact between the thermocouple and the fuel specimen as the graphite became fragmented.

Included in Figure 5 is an oscillograph record from one of the pulsed irradiations for CEN-73. This record gives an indication of the time scale of the experiments and also shows evidence for a time lag in the thermocouple. The slope of the temperature-time curve corresponds to a heating rate, during the main portion of the pulse (for the most energetic CEN-73 transient), of about 12,000°C/sec for the fuel specimen. All of the pressure records were smooth (no explosions) with the exception of CEN-75 and 76, which gave somewhat erratic traces that were attributed to failure in the circuit of the unbonded, strain gage pressure transducers. The maximum pressure rise observed was 220 psi for the final CEN-73 transient, which had an energy input of 550 Mw-sec.

After having obtained the energy-temperature data in CEN-73, other transients were conducted at various integrated power levels on short periods to determine the physical effect of such nuclear bursts. Relative "fast" periods, in the neighborhood of 100 msec, were used. At the highest energy transient (890 Mw-sec), the temperature rise of the stainless steel autoclave was 132°C.

Photographs were made of the fuel specimen to show the changes in geometry; these changes are shown in Figures 5 through 8. It is evident that each transient of 390-Mw-sec energy input, or more, was energetic enough to cause destruction of the original cylindrical fuel element under the conditions of the experiment.

Macrographs were taken of some of the particles to show the details of their shapes and sizes. Sieve-screen analyses were made on the fragments and particles formed. The particle size distributions and macrographs are also shown in Figures 5 through 8 for those specimens which had undergone destruction.

Figure 5

TRANSIENT CEN-73

GRAPHITE FUEL SPECIMEN

PHOTOGRAPH OF FUEL SPECIMEN
AFTER IRRADIATION

Energy input: several transients

MACROGRAPHS OF SOME PARTICLES

Fines

Fragments

Scale
100 mils

OSCILLOGRAPH RECORD

PARTICLE SIZE DISTRIBUTION

Figure 6
TRANSIENT CEN-74
GRAPHITE FUEL SPECIMEN

PHOTOGRAPH OF FUEL SPECIMEN
AFTER IRRADIATION

Energy input: 390 Mw-sec

MACROGRAPHS OF SOME PARTICLES

Fragments

Fines

Scale
100 mils

PARTICLE SIZE DISTRIBUTION

Figure 7
TRANSIENT CEN-75
GRAPHITE FUEL SPECIMEN

PHOTOGRAPH OF FUEL SPECIMEN
AFTER IRRADIATION

Energy input: 650 Mw-sec

MACROGRAPHS OF SOME PARTICLES

Fragments

Scale
100 mils

Fines

PARTICLE SIZE DISTRIBUTION

Figure 8
TRANSIENT CEN-76
GRAPHITE FUEL SPECIMEN

PHOTOGRAPH OF FUEL SPECIMEN
AFTER IRRADIATION

Energy input: 890 Mw-sec

MACROGRAPHS OF SOME PARTICLES

Fragments

Fines

Scale
100 mils

PARTICLE SIZE DISTRIBUTION

The mean diameter of the particles produced by the destructive transients was computed from the relationship

$$d_{sv} = \frac{1}{\sum_{i=1}^N (W_i/d_i)},$$

where d_{sv} = Sauter mean surface-to-volume diameter,

d_i = average diameter of i_{th} size group

W_i = fraction by weight of particles in the i_{th} group, and

$\sum_{i=1}^N$ = summation over N size groups.

The numerical values for W_i and d_i are shown as the ordinate and abscissa, respectively, on the histograms of the particle-size distributions. The results of these measurements and calculations of mean particle size are also summarized in Table 1.

Comparison of the data on mean diameters with the integrated power of the reactor pulse led to the conclusion that there is a trend toward smaller particles as the burst becomes more energetic. This is shown graphically in Figure 9. It is postulated that the relatively large value of 62.4 mils for CEN-73 was caused by fragmentation of the fuel at one of the lower energy bursts, and that the additional, more energetic transients caused only negligible changes in geometry. Since the original fuel cylinder had a diameter of 0.5 in. or 500 mils, it is apparent that the specific surface area of the fuel has increased by a factor of about 25 as a result of the fragmentation during transients CEN-75 and 76.

Figure 9
CORRELATION OF MEAN PARTICLE
SIZE OF SHELF-TYPE GRAPHITE
FUEL SPECIMENS AS A FUNCTION
OF THE ENERGY OF THE TREAT
REACTOR PULSE

By noting the physical changes in the fuel pin as a function of the energy of the reactor burst, it is evident that the threshold for destruction of these unclad, graphite fuel specimens is between 250 to 350 Mw-sec, which corresponds to a peak fuel temperature of 1700°C to 2000°C during a fast transient starting from 25°C with the fuel in a nonflowing, helium environment.

Absolute gamma counting of the gas samples from the autoclaves (taken into evacuated glass bulbs), with a sodium iodide crystal detector, showed the presence of xenon-133 as the main radioisotope; about one percent of the total activity came from iodine-131. By integrating under the 80-keV γ peak of xenon-133, the number of microcuries of xenon-133 was determined. From these data (Table 1) on the amount of xenon-133 found in the gas inside the autoclave, and from the number of fissions in the fuel specimen, it was computed that most of the fission product xenon was released from the fuel during the high-temperature transients (CEN-73 through 76); however, for CEN-79 and -80, in which the fuel remained essentially intact, only a few percent of the xenon was released.

IV. CONCLUSIONS

1. It was established that the TREAT reactor is a useful tool for the study of the behavior of graphite (Nerva) fuel specimens.
2. For shelf-type fuel specimens, consisting of graphite plus partially hydrolyzed uranium carbide, a disintegration threshold temperature between 1700 and 2000°C was found for reactor periods in the neighborhood of 100 msec. (Future experiments will establish this threshold more precisely.)
3. Transients which caused peak fuel temperatures lower than 1700°C caused only a few percent of the xenon-133 to be released while the fuel specimen remained intact. Peak temperatures greater than approximately 2000°C resulted in destruction of the fuel pin into fragments and particles; particle size distributions showed a decrease in the mean size as the energy of the transient increase. A mean particle diameter (surface-to-volume) of 22 mils can be attained (compared with the original 500-mil fuel diameter) with a reactor energy of 890 Mw-sec. In the destructive transients, from 44 to 73 percent of the fission product xenon-133 was released. By comparing the activity distribution data with the particle size distribution histogram, it was observed that the relative fission product release was greater from the larger particles than from the smaller particles (see Figure 10). One suggested explanation of this phenomenon is that the larger particles remained at a higher temperature for a longer time relative to the smaller particles.

Figure 10

RELATIVE AMOUNT OF FISSION PRODUCT
ACTIVITY RETAINED IN PARTICLES AS A
FUNCTION OF THE PARTICLE SIZE

V. FUTURE PLANS

The parameters to be varied and studied in this TREAT program may be classified as follows:

1. fuel
 - a. fully carbided
 - b. shelf (air stored)
 - c. maximized UO_2
 - d. coated (clad) holes
 - e. constrained (confined fuel specimen)
 - f. at high temperature at start of transient
 - g. preirradiated
2. environment
 - a. helium
 - b. water (to simulate ocean immersion)
 - c. vacuum
3. reactor transient
 - a. total energy input (integrated power)
 - b. rate of energy input (period or run time)
4. type of capsule
 - a. stainless steel autoclave
 - b. container with a window to permit high speed photography during the transient.

From these experiments, information will be obtained on the following points:

5. mechanism of destruction of fuel
6. fission product release
7. physical changes (stability of fuel, threshold of destruction)
8. particle size distribution of fragmented fuel
9. transient temperatures and pressures
10. chemical reactions

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