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PRODUCTION TEST IP-725, SUPPLEMENT A
INCREASED GRAPHITE TEMPERATURE LIMIT
F REACTOR

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HANFORD ATOMIC PRODUCTS OPERATION
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PRODUCTION TEST IP-725, SUPPLEMENT A
INCREASED GRAPHITE TEMPERATURE LIMIT
F REACTOR

I. OBJECTIVE

The objective of Production Test IP-725¹ was to investigate the reaction rate between graphite and carbon dioxide in F Reactor at a temperature 50 C higher than that currently permitted by Process Standards. After approval of the test, the temperature limit was increased from 650 to 700 C. After approximately three weeks of operation with the higher temperature limit, the reactor was shut down and the graphite oxidation monitor samples which had been inserted for this test were discharged and measured. It is believed that sufficient data have now been obtained from the first phase of the test to warrant investigation at an even higher temperature.

It is therefore the objective of this supplement to raise the graphite temperature limit at F Reactor by an additional 50 C to a limit of 750 C.

II. BASIS AND JUSTIFICATION

The basis for the present Process Standards graphite temperature limit of 650 C and the basis and justification for increasing the temperature limit to 700 C on a test basis at F Reactor are described in Production Test IP-725¹ and will not be repeated here. The basis and justification for this supplement stems from the desire to establish an upper limit to graphite temperature with respect to the graphite-carbon dioxide reaction, i.e., to determine the temperature limit for which prolonged operation of the reactor would not be recommended because of excessive oxidation of the moderator stack with carbon dioxide.

The preliminary results of the first phase of this test program are summarized in the following table and compares pertinent data between the 650 and 700 C temperature limit:

1. A. Russell. Production Test IP-725, Increased Graphite Temperature Limit - F Reactor, RL-REA-14. December 1, 1964. (Secret)

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	<u>Temperature Limit 650 C</u>	<u>Temperature Limit 700 C</u>
Graphite stringer temperature limit	735	790
Actual maximum stringer temperature observed (average)	730	775
Average maximum temperature in oxidation monitor samples	~705	~750
Approximate maximum oxidation rate in oxidation monitor samples	1/2-1%/KOD	3-4%/KOD
Approximate maximum oxidation rate in reactor stack	0.2%/KOD	0.4%/KOD
Concentration of carbon monoxide in reactor gas atmosphere	~ 0.7-0.8%	~ 1.3%
Concentration of helium in reactor gas atmosphere @ 2000 Mw	75%	50%
Average daily helium loss	~ 14,000 ft ³ /day	~ 7500 ft ³ /day

As described in the reference document, the approximate 50 C increase in graphite temperature was expected to result in an approximate eight- to ten-fold increase in oxidation rate. Measurements of the oxidation monitor samples, however, showed only an approximate five-fold increase in rate while the oxidation rate in the bulk of the stack (as indicated by the concentration of carbon monoxide) only doubled. It is postulated that the discrepancy between the observed and anticipated changes in oxidation rate may be explained by the relative contribution of the thermal and radiation induced reaction between graphite and carbon dioxide.

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As seen in the above conceptual illustration, the reaction of graphite with carbon dioxide is activated by two sources; temperature and radiation. The radiation induced reaction results from the dissociation of carbon dioxide into carbon monoxide and oxygen with the subsequent attack of the graphite by oxygen. Little is known about the kinetics of this reaction other than the reaction rate is relatively low and only slightly temperature dependent. The thermally induced reaction rate, however, is an exponential function of temperature. At low temperature, the thermal reaction rate is insignificantly low but increases with temperature until some crossover region is reached at which point the thermally induced reaction predominates. The crossover point at which the thermal reaction becomes more pronounced than the radiation induced reaction is not known exactly but is thought to be in the range of 650 C. Thus, a given change in graphite temperature below this value has relatively little effect on oxidation rate because the rate is controlled primarily by the radiation intensity rather than temperature. Above this crossover region, however, the same change in temperature has a much more pronounced effect, and the observed oxidation rate increases exponentially with temperature.

The observations made at F Reactor during this test are believed to reflect this behavior. The oxidation monitor samples are placed in uncooled graphite channels which operate at a temperature as much as 80-90 C higher than the graphite in the rest of the stack. Prior to this test, these samples operated at a maximum temperature which averaged approximately 700 C.

During the test, the average maximum sample temperature was increased to approximately 750 C. This resulted in an approximate five-fold increase in oxidation rate. These observations suggest that prior to the test, the samples were operated near the crossover region, and the increase in temperature drove the samples into the oxidation range where the thermally controlled reaction predominated. The maximum temperature in the stack filler blocks prior to the test averaged 600-650 C and was probably below the crossover region and therefore in a temperature range where the oxidation rate with carbon monoxide was primarily controlled by the radiation induced reaction. During the test when this temperature was increased to 650-700 C, the oxidation rate, as indicated by the carbon monoxide concentration in the gas system, increased only about twofold. This suggests that prior to the test the reaction rate had been controlled primarily by the radiation induced reaction and that during the test, the reaction rate had been driven up to or perhaps slightly beyond the crossover region.

Another factor which tends to prevent the oxidation rate from increasing as much with temperature as would be predicted from the laboratory data is the concentration of carbon monoxide in the gas system. As seen in the following conceptual illustration, carbon monoxide acts as an inhibiting agent to reduce the graphite-carbon dioxide reaction rate.

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In a recirculating gas system, the graphite is continually being re-exposed to the same atmosphere. Thus, the carbon monoxide, which is continuously formed as the reaction product, builds up in the recirculating system until some equilibrium condition is established. If the initial temperature were high enough to be in the range where the thermally controlled reaction were pre-dominate, the reaction between graphite and carbon dioxide at that temperature would generate carbon monoxide and an equilibrium would eventually be established between the amount of carbon monoxide being generated (a measure of the oxidation rate) and the amount of carbon monoxide being lost by gas leakage. If the temperature were increased 50 C, for example, with no change in the carbon monoxide concentration, the reaction rate would be expected to increase approximately 8-10 fold.

In practice, however, the additional carbon monoxide which would be generated by the increased reaction rate at the higher temperature would further inhibit the reaction and something less than an 8-10 fold increase would be expected. The exact magnitude of the increase would, of course, depend upon the value of the new equilibrium concentration of carbon monoxide established in the recirculating gas system.

It is possible to combine the variables of reaction rate, temperature, rate of carbon monoxide generation, and rate of carbon monoxide loss through leakage to estimate the equilibrium values of reaction rate and carbon monoxide concentration in the recirculating gas system. Detailed discussion and calculation of the interaction of these parameters is beyond the scope of this report; however, for the 50 C increase in graphite temperature proposed for this supplement, it is expected that the oxidation rate in the stack filler blocks will increase to approximately 2%/KOD, and the carbon monoxide concentration will increase to approximately 3-5%.

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Several points were mentioned in the reference document regarding the ultimate application of the information obtained from these higher temperature tests. It is probably worth repeating that these tests are intended to establish an upper limit to graphite temperature with carbon dioxide as a major component of the gas atmosphere and even if it is concluded that the current Process Standards temperature limit could be raised from this standpoint, other considerations besides the reaction with carbon dioxide may dictate the limit to graphite temperature. For example, it is questionable whether the helium saved by adding carbon dioxide to increase graphite temperature would offset the cost of the additional metal throughput required to maintain the Pu-240 within the present product specifications. Also, the long-term effects of the reaction between water vapor and graphite at these higher temperatures is uncertain. Thus, the data from these tests will provide information on only one of the parameters which go into establishing an operating graphite temperature limit.

III. TEST DETAILS

A. Procedure

1. Upon approval of this supplement, the graphite temperature limit at F Reactor will be increased by 50 C. For the duration of this test, the graphite stringer temperature limit will be 850 C (corresponding to a maximum graphite filler block temperature of 750 C). It is estimated that this temperature increase can be achieved by a reduction in helium percentage of approximately 15-20 per cent.
2. The change in the relative proportions of helium and carbon dioxide in the gas atmosphere which will be associated with this test will result in an increase in the bulk gas density and therefore a change in the pressure difference between the plenum chamber and the outside air on both the front and rear faces of the reactor. When the reactor gas is lighter than air, such as with the present gas composition, the positive pressure which is currently measured in the lower region of the plenum chamber with respect to the outside atmosphere becomes even more positive with increasing elevation, thus assuring that gas leaks out of the system rather than air leaking in. When the reactor gas is heavier than air, as with 100 per cent carbon dioxide, the gas pressure inside the plenum chamber decreases more rapidly with increasing elevation than does the outside air. Thus, a slight positive pressure measured at the bottom of the reactor could reverse to a slight negative pressure at the top. It may therefore be necessary to slightly increase the indicated unit gas pressure to insure that this change in gas density does not cause air to leak into the system. The ground rules for controlling air leakage should continue to be to hold the reactor gas pressure to that value which maintains the nitrogen concentration within the 0.1 per cent limit recommended in the reference document. It is expected that only a slight pressure increase, if any, will accomplish this objective.

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3. The increase in graphite temperature is expected to decrease product buildup in the irradiated fuel and increase the amount of Pu-240 isotope in the product. An additional increase of 0.2 per cent absolute in F Reactor Pu-240 content should be assumed for the duration of this test when blending future product from F Reactor in the 200 Areas with that from other reactors².
4. Carbon monoxide and hydrogen concentration limits during this test are discussed in the Hazards and Limits sections.

B. Instrumentation

1. Graphite oxidation monitoring samples presently in tube channels 1067 and 3580 will be charged out and replaced with new samples.
2. The presently installed chromatograph is calibrated to measure carbon monoxide concentrations up to five per cent. Although there is the prospect that this concentration could be exceeded during this test, the present chromatograph can be adequately reranged and calibrated with present equipment.
3. The increase in graphite temperature will increase the reaction rate between graphite and the water vapor in the gas atmosphere. This reaction can be monitored by measurement of one of the reaction products, hydrogen. A hydrogen analyzer with adequate range and sensitivity was installed and used during the initial temperature increase and will again be used during this test.

C. Data Required

1. Graphite oxidation which has occurred during the test will be measured at the conclusion of the test by analysis of the oxidation monitoring samples.
2. Oxidation during the test will be monitored by analysis of the reaction products, carbon monoxide and hydrogen, in the reactor gas system. The continuously recording chromatograph and hydrogen analyzer will be used for this purpose.
3. Gas loss information will be available from operational records.

IV. HAZARDS

There is always the prospect that the oxidation rates during the test may be higher than anticipated. In addition, data on the graphite-water vapor reaction, as it applies to stack geometry, is very limited. There is, therefore, the hazard that at the higher temperature authorized for this test,

2. Personal Communication with A. D. Vaughn, Operational Physics, Research and Engineering Section.

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these reactions may proceed at a rate which is unacceptable for prolonged reactor operation. One of the primary purposes for monitoring the carbon monoxide and hydrogen reaction products in the gas system is therefore to provide early warning of excessive oxidation rates if they should exist.

The additional stack oxidation which will occur during the test period will be arbitrarily limited to one per cent, corresponding to an oxidation rate limit of ten per cent per thousand operating days during the three-month test period. The carbon monoxide and hydrogen concentrations in the reactor gas system which would result from this rate are 15-20 per cent and 8-12 per cent, respectively, and will serve as a guide until the oxidation monitor samples can be removed and analyzed.

The actual permissible carbon monoxide and hydrogen concentration limits, however, will be revised downward from these values for the duration of this test for another reason. Both carbon monoxide and hydrogen are explosive in air if present in sufficient quantities and thus represent a potential safety hazard. The explosive concentration range of carbon monoxide in air is 12-75 per cent, while the range for hydrogen is 4-75 per cent. When both gases are present in the same mixture, however, the explosive concentration is altered and depends upon the relative proportions of each gas. Figure 1 shows the maximum permissible concentrations of carbon monoxide and hydrogen which will insure that no explosive mixture results. It is this curve which should be used as the limit to carbon monoxide and hydrogen during this test. This curve is considered conservative in that even if these percentages were observed, the leakage of gas out of the reactor or the leakage of air into the system would result in sufficient dilution that the mixture would no longer be flammable. The values shown in Figure 1 are not expected to restrict the temperature increase authorized by this supplement except that in the event of a large water leak, sufficient carbon monoxide and hydrogen could be liberated that corrective action would be necessary. If the limits are exceeded, the graphite temperature should be reduced and the gas pressure increased until the carbon monoxide and hydrogen concentrations are within the limits shown.

V. PROCESS STANDARDS

As stated in the reference document, all Process Standard limits with regard to over graphite temperature limit provisions and inlet dew point requirements will remain in effect except those specifically exempted by PITA-26-I, Supplement C.

The reduction in helium percentage is expected to result in an increase in biological shield temperature of not more than 30 C. Although 30 C is not expected to increase the shield temperature to the 160 C limit which would require reactor shutdown, it could drive the temperature up into the 110-160 C range. Process Standards recommends charging fringe poison to maintain shield temperatures below 110 C; however, for the duration of this test, maximum shield temperatures in the range of 110-160 C will be considered acceptable without charging additional fringe poison.

VI. LIMITS

The various temperature and gas system limits for this supplement are summarized as follows:

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1. Graphite temperature limit - 750 C in graphite filler blocks
(corresponds to 850 C on front-to-rear thermocouple stringers)
2. Over graphite temperature limit - Process Standards, Section C-040
3. Normal nitrogen limit - 0.1%
4. Normal oxygen limit - 0.02%
5. Startup oxygen limit - 1% when power level exceeds 1 Mw
6. Carbon monoxide limit - Variable with hydrogen concentration - See Figure 1
7. Hydrogen limit - Variable with carbon monoxide concentration - See Figure 1
8. Moisture content - Process Standards, Section B-010
9. Biological shield temperature - 160 C

VII. TEST DURATION

This test is expected to last approximately three months but may be terminated early:

1. When it is believed that meaningful oxidation results can be obtained from the oxidation monitoring samples.
2. If the concentrations of carbon monoxide or hydrogen cannot be controlled within the limits shown in Figure 1.
3. If biological shield temperatures exceed the current Process Standards limit of 160.
4. Upon mutual concurrence of the Manager of F Processing Operation, Process and Reactor Development, and Process Technology.

A Russell

Reactor Engineering
Process and Reactor Development

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VIII. RESPONSIBILITIES

All responsibilities noted in the parent document shall remain unchanged.

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Figure 1 - Limit to Carbon Monoxide and Hydrogen Percentages

Example: If carbon monoxide concentrations were 6%, hydrogen concentration should not exceed 2%.

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