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MTR REFLECTOR GRAPHITE AFTER 13 OPERATING YEARS*

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INTRODUCTION

The Materials Testing Reactor has now been in operation for over 15 years. Limited surveillance of the reflector graphite condition has been carried on periodically during this time, and particularly after the Windscale incident of inadvertent energy release. The most complete or detailed analysis was made several years ago and forms the basis of this report. The object was to assure that continued operation of the MTR was safe. The test procedures used were conventional, designed to give a reasonable picture of the graphite condition.

The design of the MTR as seen in Figure 1 reflects the adverse experience with graphite in the first production reactors at Hanford. No reflector graphite was placed next to fuel. The graphite zone next to the tank, nearest the fuel, was filled with one-inch graphite balls to avoid distortion of experimental facilities to discharge the balls when necessary and replace them with fresh material. Solid bar graphite was used only outside this inner zone, i.e., where the anticipated flux would be low. An expanded view of the center horizontal cross section and experimental facilities is shown in Figure 2.

In spite of a limited program of graphite irradiation damage studies at the MTR, and the stated purpose to monitor the reflector graphite, comparatively little was done during the first five years of operation. A few stored energy measurements were made of graphite from certain plugs of the VG experimental holes and found to be lower than anticipated. A few capsules full of machined graphite specimens were placed for an indefinite irradiation period into VG-18,

one of the graphite zone facilities of highest fast flux. Some of these were measured in 1956. After the Windscale incident caused some excitement others of these were measured also. Further samples were taken from HG-5 and -6 graphite plugs for analysis in 1958 and from VG-10 in 1959. A rather detailed analysis of the safety factors were made at this time and reported in IDO-16656. Results indicated that for at least 5 years, an accidental spontaneous energy release would not result in physical damage to the reactor structure.

Thus, the latest check on the condition of the MTR graphite was carried out in 1964. Since a more complete and comprehensive study was made then, the present report is largely the result of this investigation. A relatively large number of graphite balls or pebbles were taken from various levels through the facilities VG-12 and VG-20, as shown in Figure 3. Since sample balls were retrieved as each facility liner was withdrawn it was felt that the positions from which the samples came were known with reasonable precision. In addition activated balls with thermocouples were placed for future monitoring during this operation.

This paper then is a summary of the tests made on the MTR graphite, particularly during the latest series of measurements.

MEASUREMENTS

Fast flux measurements (above 1 MeV) were made with nickel wire in the VG facilities 7, 12, 13, 33, and 41, using the threshold reaction $\text{Ni-58}(n,p)\text{Co-58}$. In Figure 4 a plot of the data shows a logarithmic relationship of intensity as a function of distance from the active core. The integrated fast flux at the vertical maximum was about 2×10^{19} nvt at the time of sampling. This is, of course, a fast flux considerably below that found in graphite power or production reactors.

The most important quantity to determine from the standpoint of safety was the accumulates stored energy releasible at relatively low temperatures. Sampling of graphite from the pebble zone of the MTR reflector was done with a specially designed tool through VG-10 and directly from the bottoms of the holes of VG-12 and VG-20, as stated previously, while the liners were being withdrawn. Thus representative samples were obtained near the tank along vertical traverses on different sides of the reactor.

Figure 5 shows the type of data curves obtained in the procedure for measuring the stored energy release. The graphite ball containing an imbedded thermocouple was placed in a specially designed oven whose temperature was held constant at 200°C. An initial rise in temperature occurred to above that of the oven, followed by a gradual cooling until the ball temperature eventually came into equilibrium with the oven. A post-anneal run of the same type, during which time no stored energy was released gave a value of the temperature differential needed to calculate the results. An energy release rate curve calculated from these data is shown in Fig. 6. Combining the results from sample balls from each of the individual facilities gave an indication of a traverse in the accumulated energy released at this anneal temperature, as shown in the next three figures.

Figure 7 gives data from VG-12 balls taken in December 1963 at the end of 112,550 MWD, with a maximum of about 65 to 70 cal/g. The scatter of data points may be due in part to the uncertainty in the sampling procedure. However, some evidence also indicated a spontaneous energy release in individual balls in the reactor without triggering a release in adjacent balls. Some samples which showed little energy released at 200°C did, however, show a relatively large amount in total heat of combustion

measurements. The relative C^{14} activity also shown in Fig. 7 indicated a total irradiation commensurate with the position even though in some cases the stored energy released was small. X-ray results are also given in per cent change of crystallite c_0 spacing on the same figure and are in essential agreement with the stored energy results as to total fast neutron dose these effects represent. The maximum in the c_0 spacing change occurs at a slightly different position on the vertical from that of stored energy due to different buildup and anneal mechanisms of the two phenomena.

Figure 8 shows the corresponding data from VG-20 with a maximum stored energy of about 70 cal/g. The irradiation history of the MTR was the same as for VG-12. Figure 9 shows a plot of data from VG-10. The balls were taken in April 1959 after only 61,353 MWD of operation.

Another quantity measured was the ignition temperature at which oxidation would be sustained in air, i.e., at which the time-temperature curve showed an inflection. Data are summarized in Table I. The samples were heated with an induction heater in an environment of uniform air flow, maintained at a rate of 10 liters/minute in a 1.1" diameter tube. The point at which the time-temperature curve showed an inflection was taken as the ignition temperature. Unirradiated balls ignited at about 830°C whereas the temperature for irradiated balls was about 100°C lower. Statistics were not very good. However, this was still 300°C higher than the maximum temperature anticipated from any spontaneous energy release.

Table I

GRAPHITE IGNITION MEASUREMENTS-I

Sample Location	No. Samples	Avg. Temperature °C of Inflection
Virgin	3	830 $\pm$ 10
VG-12 Center	3	750
VG-12 Top	2	700
VG-20 Center	2	790
Avg. all Irradiated Samples		748 $\pm$ 50

Table II summarizes data of a second series of ignition tests in which different induction heating equipment was used and the ignition temperature was defined as the point at which a blue flame appeared when viewed through a blue filter (Corning 4308). Results show a temperature appreciably higher than the other set. Furthermore, the flame point of the irradiated specimens is higher than that of the virgin balls instead of lower. However, the fluctuations are rather large and statistical uncertainties overlap in most cases.

Table II

GRAPHITE IGNITION MEASUREMENTS-II

Sample Location	Number of Measurements	Avg. Flame Point Temp. °C
Virgin	10	1138 $\pm$ 85
Top of MTR Ball Zone		
VG-10	9	1228 $\pm$ 56
VG-12	10	1294 $\pm$ 61
VG-20	11	1201 $\pm$ 64
Center of MTR Ball Zone (1964)		
*VG-12	9	1244 $\pm$ 69
*VG-20	11	1209 $\pm$ 101

*No Ignition on One Sample at > 1320°C (Not included)

The final type of measurement was that of weight loss in individual balls, pictured in the curve of Figure 10. Each ball retrieved was weighed and the average compared to the average of a large number of virgin balls. The change in weight is statistically significant although variations in individual balls were rather large. Included are data of balls recently taken from the top of the pebble bed shown as a triangular point. These samples show no weight loss since they were out of the radiation zone. During operation the temperature is about 40°C here compared to 60 to 90°C in the irradiation zone. Thermal flux from C^{14} activity measurements show an attenuation of at least a factor of 20, while the fast flux is essentially zero.

The overall loss rate is summarized in Table III. The rate of material loss is small, being only about $0.20 \pm 0.05\%$ per 10^4 MWD of MTR operation (about one year). If oxidation is the responsible mechanism the loss in graphite would occur as CO or CO_2 and should be temperature and possibly neutron flux dependent. Broken down into data from the center ± 12 inches from the MTR core center plane and that outside this region shows a slight difference-- $0.24 \pm 0.05\%$ and $0.15 \pm 0.05\%$ respectively--although the statistics make it of doubtful significance. If indeed a temperature dependent phenomenon is being observed the temperatures involved are much lower than given for other published data. The calculated rate at which C^{14} is discharged to the atmosphere via the stack gas is about 4×10^{-8} μc per cm^3 compared to a published maximum permissible level of about 10^{-6} $\mu\text{c}/\text{cm}^3$.

Table III

MTR GRAPHITE WEIGHT LOSS

<u>Region</u>	<u>Weight Loss % per 10^4 MWD</u>
Irradiation Zone	$0.20 \pm .05$
Center $\pm$ 12 inches	$0.24 \pm .05$
Outside $\pm$ 12 inches	$0.15 \pm .05$
Top of Pebble Bed	None Detectable
C^{14} discharged to atmosphere approximately 4×10^{-8} $\mu\text{c}/\text{cm}^3$ during normal MTR operation 1964	

ACCIDENT ANALYSIS

Since the source of a potential accident damaging to the MTR is primarily a spontaneous release of stored energy it is of interest to know what potential thermal sources exist that might trigger such a release. Temperature traverses are given in Figure 11 for the region of maximum neutron flux and gamma heat generation in VG-12 and VG-20. These data indicate a lack of uniform air flow. The maximum gamma heat supplied by the MTR operating at 40 MW is about 1 watt per pebble which would result in a temperature rise of 3 to 4 C per minute. The normal cooling of the graphite in the pebble zone after a scram is pictured in Figure 12. Air temperature is reached in 20 to 25 minutes. Adding gamma heat to the anticipated contribution from stored energy gives the temperature transients during several conceivable abnormal conditions as shown in Figure 13. An analysis shows that about 18 minutes is available before a power reduction will not control the temperature if one of two blowers should fail. On the other hand, if a step power change of 50% should occur while maintaining both blowers, the critical point is not reached for over 30 minutes. This condition would

normally scram the reactor immediately. Either case allows ample time to initiate appropriate safety measures.

As a final calculation, assuming an adiabatic release of the stored energy by any conceivable means, the temperature rise was estimated assuming alternately that several different regions of the graphite absorbed all the heat. Figure 14 shows such calculations made in 1959 with an extrapolated value to 5 and 10 years later (based on rather meager and uncertain data) showing a possible danger point would be reached in five years. The measured values in 1964 showed the previous extrapolation to be greater than the actual value by a factor of two. New extrapolations give reasonable assurance of no excessive temperature rise following a spontaneous release until at least 1970. A controlled energy anneal program has been outlined if an anneal is deemed desirable (IDO-16656). The anneal operation in place would be much more economical than to carry out such a program outside the reactor or to replace the entire batch of 720,000 graphite balls.

SUMMARY AND CONCLUSIONS

The stored energy releasible at low temperatures in the MTR graphite reflector is approaching saturation and does not appear to present a serious risk to continued operation for any credible operational failure. The approach to any condition that conceivably would initiate a spontaneous energy release is sufficiently gradual that adequate corrective measures can be taken. The buildup of radiation damage is consistent with that expected from the operating temperature and measured fast flux. Good agreement is found with previous work to which the present measurements can be compared.

The maximum temperatures reached in spontaneous release would still be several hundred degrees centigrade below ignition point. Slow oxidation was

measured with statistical significance. The temperature and fast neutron flux levels as well are much lower than that at which weight loss rate has previously been measured.

REFERENCE

E. Fast, E. O. Smith, and J. D. Ford, "A Proposal for the Release of Stored Energy in the MTR Reflector Graphite", IDO-16656 (1959).

MTR VERTICAL E-W CROSS SECTION

Fig. 1

MTR HORIZONTAL CROSS SECTION

Fig. 2

SAMPLE AND THERMOCOUPLE LOCATIONS

Fig. 3

INC-C-9748

Fig. 4

ANNEAL of IRRADIATED GRAPHITE

Fig. 5

INC-B-9756

ENERGY RELEASE RATE CURVE

Fig. 6

Fig. 7

Fig. 8

INC-B-9761

Fig. 9

INC-B-9758

GRAPHITE BALL WEIGHT LOSS WITH MTR OPERATION

Fig. 10

INC-B-9754

MTR PEBBLE BED TEMPERATURE & GAMMA HEAT PROFILES

Fig. 11

MTR GRAPHITE PEBBLE BED TEMPERATURE AFTER REACTOR SCRAM

Fig. 12

MTR PEBBLE BED TEMPERATURE TRANSIENTS

Fig. 13

MTR GRAPHITE TEMPERATURE AFTER ADIABATIC STORED ENERGY RELEASE

Fig. 14