MASTER

THE PEBBLE BED REACTOR PROGRAM

Summary of the First Capsule Irradiation

January 14, 1959

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An irradiation program was conducted during the past 5 months which had the primary objective of determining the structural integrity of various types of uranium graphite fuel elements that could be used in a Pebble Bed Reactor.

I Specimens

All specimens were nominal 1 1/2 in. dia. spheres of graphite fueled with 4.75 gms. of 93% enriched uranium in the form of uranium dioxide. Two specimens of each of three types were irradiated.

Impregnated: Graphite spheres were machined from extruded AGOT stock (National Carbon Co.) and impregnated with an aqueous uranyl nitrate solution. The uranyl nitrate was converted to UO_2 by baking up to a maximum temperature of $800^\circ C$. The impregnation work was done by the Sylvania Corning Nuclear Corp.

Admixture: A mixture of high fired UO_2 shot (Mallinckrodt Chemical Works) in the size range of 105 to 149 microns was blended with type 2301 graphite powder filler and a coal tar pitch binder. The mixture was first molded and then baked up to a maximum temperature of $2500^\circ F$. The uranium remained in the form of UO_2 . These specimens were made by the National Carbon Co.

Lumped: A 1 in. dia. "green" sphere was first formed by pressing a mixture of 105 to 149 micron UO_2 , a phenolic resin coke filler (Type 79L) and a phenolic resin binder (Type BV-1600). The 1" sphere was then placed inside a 1/4" thick unfueled spherical shell which consisted of AGOT graphite flour filler and a phenolic resin binder (Type BV-1600). The fuel

element was then molded under 10,000 psi pressure and then baked at a maximum temperature of 2000°F. These specimens were made by the Battelle Memorial Institute.

II The Irradiation

The encapsulation, irradiation, and examination of the specimens were performed by the Battelle Memorial Institute. The specimens were mounted in graphite cylinders which were hollowed out to receive the spherically shaped specimen. Loosely packed graphite powder was placed in a 30 mil. annulus between the specimen and the graphite cylinder in order to enhance heat transfer. Two such graphite cylinders containing their specimens were mounted in a finned steel inner capsule. The fin thickness and spacing were selected to produce a specimen surface temperature of approximately 1500°F based on an anticipated heat generation rate of 2 KW per specimen. No auxiliary heat was provided. The three finned inner capsules were mounted in an outer capsule which was 2 1/2" OD by 17 in. long. 12 thermocouples were located throughout the capsule. Two were located just within the surface of the uppermost specimen, the remainder were located throughout the various graphite support cylinders adjacent to the graphite powder annuli. Dosimeter wires were located around each specimen.

The irradiation was performed in the Battelle Research Reactor, a 1 MW swimming pool reactor. The capsule was located in a central hole having a maximum unperturbed thermal neutron flux of nearly 4×10^{14} n/cm²-sec. The capsule was inserted on October 22, 1958 and removed on Dec. 1, 1958. The total time at full reactor power was 792 hours. Heat generation rates in the specimens were calculated from the observed temperature readings and the thermal properties of the capsule. The calculated average heat generation rates ranged from 1.7 to 1.9 KW and the calculated peak heat generation rates ranged from 1.9 to 2.3 KW. These peaks occurred for relatively short periods of time (30 to 60 hrs.), coinciding with reactor start-up periods and addition of new BRR fuel elements. Specimen surface temperatures ranged from 1200°F to 1350°F. Based on a total exposure of 1345 KWH (i.e. 1.7 KW for 792 hrs.), the probable burn-up in the specimens can be expressed in a variety of ways as follows: (a) 1.5% burn-up of U235 atoms, or (b) 0.007% of total atoms in the specimen, or (c) 10,800 MWD/Ton of uranium, or (d) 5.8×10^{18} fissions/cc of matrix in the case of the impregnated and admixed elements which had uniformly dispersed fuel throughout and 2×10^{20} fissions/cc of the central core of lumped element.

III Test Results

A. Dimension and Weight Change

		Diametral Shrinkage, %			Weight change %
		X	Y	Z	
IMPR	#13	0.847	0.82	1.088	-4.5
	#16	0.90	1.068	0.807	-2.8
ADMX	#86	0.073	0.240	0.280	+0.03
	#87	0.286	0.266	0.374	-0.08
LUMP	#11	0.320	0.263	0.267	-4.2
	#12	0.093	0.274	0.153	-0.7

The X and Y axes are perpendicular to the direction of the molding force (admix. and lumped) and the extrusion axis (impreg.) while the Z axis is parallel with the same directions. The dimensional changes give a good indication of the relative fission fragment damage to each of the three types of elements. The larger shrinkage in the case of the impregnated specimens is due to the small UO_2 particles (~ 1 micron) which permit a large fraction of the fission fragments to escape and give up their energy to the graphite matrix. The larger UO_2 particle size in the case of the admixed specimens and the lack of fuel in the outer shell of the lumped specimens are responsible for the lesser shrinkage.

In order to determine whether the weight losses were associated with radiation effects, similar unirradiated specimens were heated in argon for 8 hours at 1500°F . The resulting weight changes were:

IMPR - -0.18%

ADMX - -0.03%

LUMP - -0.61%

The weight losses of the unirradiated specimens are not of the same magnitude as for the irradiated specimens due to the shorter time at temperature. However, it appears that the weight losses can be associated with temperature effects because of the similar relative trend in each case. (i.e. lowest loss for admix.). All of the specimens were

manufactured at or above the 1500°F test temperature, so that it is unlikely that the losses are due to further volatilization of the fuel element constituents. It is more probable that the losses are due to release of adsorbed atmosphere gases, most likely water vapor.

B. Compression Tests

The specimens were subjected to an increasing compressive load to the point of failure. The load at fracture and the slope of the load-deflection curve (S, lbs/in) are shown below:

	Pre-Irradiation		Post Irradiation	
	Load	S	Load	S
IMPR	2,555 lbs.	25,900 lbs/in.	2,672 lbs.	39,100 lbs/in.
ADMX	2,410	56,000	3,712	66,000
	2,865	54,000		
LUMP	1,060	43,500	258	8,300
	(708)	(40,500)		

In the case of the pre-irradiation test of the lumped specimen, a sharp jump in the deflection reading was noted at 1060 lbs. No visible crack in the outer shell was observed. Upon reloading, the specimen broke at 708 lbs.

The compressive strength of the impregnated and admixed specimens both showed a definite increase with irradiation, a characteristic which relates to the increase hardness observed for unfueled graphite under neutron irradiation. Another measure of the greater fission fragment damage to the graphite matrix is shown by the relatively greater increase in the pseudo modulus of rupture, S, for the impregnated element. The lowered compressive load for the lumped element was probably due to a readily visible crack which had developed in the outer shell of the compression test specimen during irradiation.

C. Impact Tests

Impact tests were performed by dropping a 6 1/4 lb. weight on the specimen from successively greater heights until the specimen fractured. The results expressed as ft-lbs. at fracture are:

	Pre-Irradiation	Post Irradiation
IMPR	13.5 ft. lbs. 11.5 12.5	10.4 ft. lbs.
ADMX	7.3 11.5 11.5	8.35
LUMP	3.1 2.0	1.04

Although the pre-irradiation data showed some scatter, it can be concluded that the IMPR and ADMX elements suffered only a slight loss in resistance to impact loads. This point is of particular interest since the abrasion and compression tests had indicated an increase in hardness and modulus of rupture. Both of these irradiated specimens required approximately ten impacts to the point of failure during which time, no noticeable dusting was observed. Both of these specimens broke with a clean sharp fracture into two approximate hemispheres. The LUMP element failed at the first impact with the shell breaking into several pieces. The core piece was also fractured. Since this latter feature was not evident in the pre-irradiation tests, it was concluded that the core piece probably shrunk away from the shell during irradiation, causing successive impacts on the shell and core.

D. Abrasion Tests

Surface stability after irradiation was determined by tumbling the test specimen together with a number of dummy elements (admixture type) in an 8 in. dia. by 4 3/4 in. long rubber lined drum. The drum was rotated by a set of external rollers. Visual observation and weight loss were the criteria of abrasion resistance. Although the wide range of weight losses in pre-irradiation testing showed that this was not a good quantitative test, the post irradiation tests effectively demonstrated that surface stability was not impaired. In fact, the weight gains shown below are interpreted to mean an increased surface hardness.

The values shown are weight change in grams during the tumbling period which was 2 hours for the unirradiated samples and 10 min. for the irradiated samples.

	Pre-Irradiation	Post Irradiation
IMPR	- .0352 max	+ .0025
	- .0156 avg	- .0008
	- .0008 min	
ADMX	- .0281 max	+ .0026
	- .0088 avg	+ .0024
	- .0003 min	
LUMP	- .3081 max	- .0185
	- .0928 avg	
	- .0035 min	

E. Fission Product Leakage

Fission product gases which escaped to the inner capsule gas space were collected by puncturing the inner capsule with an O-ring-sealed puncturing device and releasing the gas to an evacuated collection system. In the case of the impregnated specimen, the results corrected to the time of capsule removal from the BRR gave the following:

$$\frac{\text{Xe 133 in gas space}}{\text{Total Xe 133 present}} = 0.005\%$$

In the case of the admixtured elements, the fission gas sample showed an extremely low activity, approximately a 10^5 lower disintegration rate than the impregnated sample. Subsequent analysis of the admixtured samples by a mass spectrometer gave the following results, expressed as percent by volume:

	<u>Sample #1</u>	<u>Sample #2</u>
Air	95%	98%
H ₂	4	trace
Kr	0.02	0.01
He	No more than normal quantity found in air	

These results indicate that there was an air leak at the puncturing device joint. Since the capsules were originally loaded with helium, its

absence indicates that the capsule gases must have first leaked to the atmosphere, and then air entered the collection system. The presence of hydrogen was due to the radiation decomposition of adsorbed water vapor, one of the factors in the weight loss results.

No results were obtained for the lumped specimens since its capsule was inadvertently punctured while opening the outer capsule.