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Design and Feasibility Study of a Pebble Bed Reactor-Steam Power Plant

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1.0 Fuel Element Technology

The Pebble Bed Reactor concept is based on the use of a spherical graphite element containing fissile and/or fertile material in the form of an oxide or carbide, heat of fission being removed by the circulation of helium through the reactor core. A reactor of this type was first proposed by Dr. Farrington Daniels (1) in 1944 and is currently being studied under the present contract as a heat source for a central station steam power plant. Other applications of the same reactor concept that are being investigated are a process heat reactor by the U. S. Bureau of Mines and a central station power reactor in Germany by Brown-Boveri & Cie, A. G., Mannheim and Friedr. Krupp.

Graphite-uranium fuel elements have been investigated in a random fashion over the past ten years, fuel being incorporated in the graphite matrix by three general methods: (1) impregnation of a graphite shape with a uranium compound followed by a conversion to the oxide or carbide form by heating; (2) admixture, or the mixing of a uranium compound such as the oxide with a graphite flour and a binder, followed by forming and baking to carbonize or graphitize the binder; and (3) lumping, where a lump or pressed shape of uranium oxide or carbide is sealed within a preformed graphite body. Immediately following the proposal by Dr. Daniels mentioned above, the Argonne National Laboratory and the Oak Ridge National Laboratory made a study of the impregnation technique (2) (3) (4). The next effort along these lines was by North American Aviation (5) who had under consideration a low power research reactor and a high temperature power reactor based on impregnating blocks of graphite with fissile material. Battelle Memorial Institute (6) initiated studies of this fuel concept in 1950, concentrating on the addition of fuel to graphite by the admixture method. This work is still being carried on at an accelerated level. During recent years both the Los Alamos Scientific Laboratory (7) and the University of California Radiation Laboratory at Livermore have done work on impregnation and work is still going on in these laboratories on the fabrication of graphite-uranium fuel elements by both impregnation and admixture. In 1957 the Commission contracted with the Sylcor Corporation to investigate the preparation of graphite-uranium fuel elements and while the greater part of their work has been on graphite techniques, they have fabricated sample lots of graphite-uranium and/or thorium elements. Great Lakes

Carbon Company are presently fabricating admixtured fuel elements for TREAT, a pulsed research reactor being built by Argonne National Laboratory, and Battelle Memorial Institute are having admixtured graphite-uranium fuel elements developed by three graphite manufacturers for the GCRE Program.

In the private field Dr. Daniels has done limited work at the University of Wisconsin on admixture and lumping; National Carbon are doing work on admixtures; Minnesota Mining and Manufacturing are doing work on lumping and high temperature coatings and The Engineering Sciences Division of American Metal Products Company are doing work on gaseous impregnation and diffusion coatings.

In the foreign field, Degussa of Frankfort, Germany have provided lumped elements for BBC-Krupp which have been irradiated at NRX in Chalk River, and the British have reported irradiation results on graphite-uranium bodies.

In general this work has been or is being done in a random or uncoordinated fashion and at a low level of effort. There is no question regarding the feasibility of fabricating fuel elements of this type but performance data is scattered or completely missing. This is because of the shortage of test facilities, the lack of priority to make testing possible and the heretofore low level of interest in the high temperature, gas-cooled reactor concept.

In the following section of this report a summary is presented of work done in this field, as outlined above, as related to the PBR concept. Since this work originated in several different locations and was based on programs with different aims, it is not necessarily a continuous picture, making it difficult to correlate the results.

1.1 Materials

The temperature dependent physical and chemical changes associated with a variety of materials that may be used in the fabrication of graphite fuel elements for the pebble bed reactor are indicated graphically in Figure 1-1. Included on this chart are graphite, fissile and fertile materials, the combination of graphite and fissile materials and some potential coating materials.

1.1.1 Graphite

In the pebble bed reactor graphite serves as the fuel element structural material and as the moderator. The graphite density will be dependent upon the method of fuel element fabrication. Adding uranium to the matrix by admixture or lumping permits the fabrication of bodies of high graphite density. Adding uranium to the matrix by impregnation necessitates a body having interconnected pores with the graphite density being dictated by the weight percent of fuel to be added.

Graphite can be manufactured from almost any organic material that leaves a high carbon residue on heating. Petroleum coke is one of the most economical and widely used of the available raw materials. The coke as received from the refinery is fired or calcined at a temperature of about 1300°C to preshrink the coke so that the volume changes during subsequent processing will be controllable.

The calcined coke is crushed and screened to yield various sizes which are mixed to form an appropriate blend. This blend is then mixed with a pitch binder. The viscosity of the pitch is strongly temperature dependent and in order to achieve a good mix, the mixing is carried out at about 165°C, at which temperature the pitch is quite fluid. The proportion of pitch to coke is selected so as to yield a mixture that is plastic enough for extrusion or molding.

After forming, this "green" product is cooled to harden the pitch binder and make handling of the product possible. The product is fired in a gas-fired furnace at about 750°C to carbonize the binder. During this process the green product is packed tightly in a coke or coke and sand mixture so that it will not distort when passing through the temperature range at which the pitch is fluid. After gas firing, the carbon product is reimpregnated with pitch to increase the density, if required, and finally graphitized at 2600°C to 2800°C.

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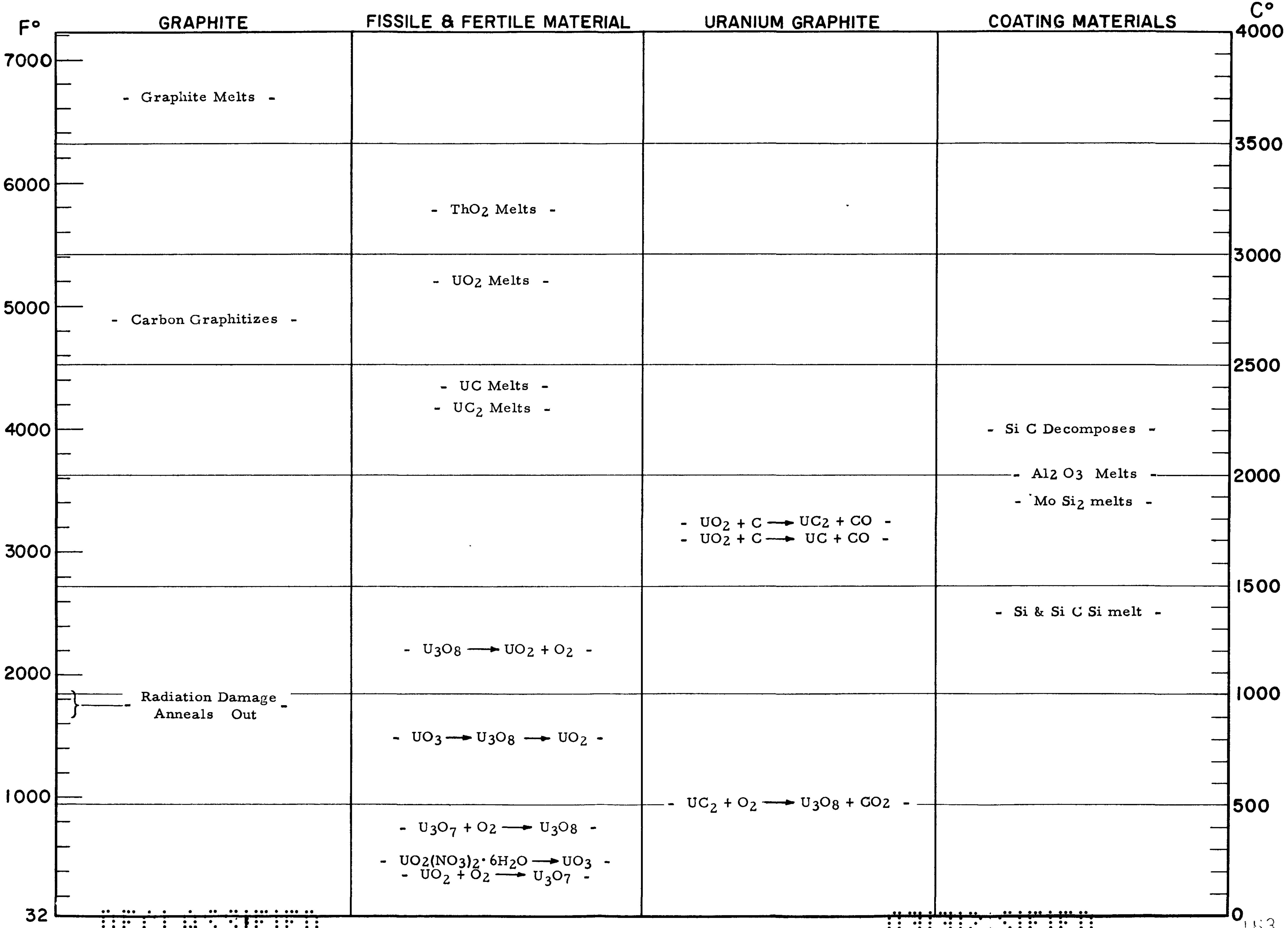
FIGURE 1-1. **UNCLASSIFIED**



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Since the coke particles tend to line up with their longest dimension parallel to the axis in extrusion and perpendicular to the force in molding, the resultant products have different characteristics, even when made from the same stock. Characteristics of graphite made by these two methods are given in Table 1-1.

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TABLE 1-1

Effect of Molding vs. Extrusion on Physical Properties of Fine-Grained Stock (8)

	Extruded	Molded
Density - gr/cm^3	1.64	1.75
Coeff. of Thermal Expansion ($x10^{-7}/°C$)		
With Grain	11	19
Against Grain	41	32
Ratio	3.70	1.68
Elastic Modulus (10^6 lbs/in^2)		
With Grain	1.84	1.39
Against Grain	0.78	0.96
Ratio	2.40	1.45
Modulus of Rupture (psi)		
With Grain	4520	4680
Against Grain	3010	3940
Ratio	1.46	1.25

In fabricating graphite-uranium fuel elements by admixture it is desirable to be able to control the chemical form of the fissile material in the final product. From inspection of Figure 1-1, it is apparent that if the oxide, for example, were mixed with calcined coke and a pitch binder and graphitized, the oxide would convert to the carbide. In order to establish some degree of control over the final product, admixtured elements are made by mixing a suitable form of graphite flour, uranium oxide and a pitch binder and carbonizing the binder. This results in a homogeneous mixture of graphite, UO_2 and carbon. This mix can also be fired at graphitizing temperature which results in a homogeneous

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mixture of graphite and UC_2. Carbonizing or graphitizing the pitch binder results in a significant difference in physical properties, as shown in Table 1-2.

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TABLE 1-2

Properties of Molded Graphite Bodies (9)

Final Baking Temperature °C	Bulk Density gr/cm^3	Thermal Conductivity Btu/Hr Ft°F	Modulus of Rupture psi		Compressive Strength psi
			w	a	
1450	1.72	12.8	-	6600	15,800
2800	1.70	26.0	6000	4680	10,070

w - With Grain a - Against the Grain

There has been a desire on the part of certain fabricators to produce a product at room temperature and one that would not soften during baking so that the bodies would have good dimensional stability during this process. As a result, various laboratories (6) (10) have developed a method of preparing a matrix with a graphite flour and plastic binder which can be mixed and formed at room temperature and then baked. High densities are obtained by this method without further impregnation with binder and the bodies have excellent high temperature properties.

The use of plastic or resin binders is of particular interest where high density bodies are required. Battelle report achieving bulk densities of 1.82 gr/cm^3 using phenol formaldehyde binder and LASL report achieving bulk densities of approximately 1.80 gr/cm^3 using a furfurol plastic binder, both without further impregnation with binder. Commercial pitch bonded bodies have a density of 1.7 to 1.8 gr/cm^3, but only after impregnation with pitch after the initial baking.

By varying the grain size of coke particles or graphite flours, the kind and amount of binders, the method of forming and the final bake time and temperature, an almost infinite variety of stocks can be produced having widely different properties. Some of the major problems in producing reactor grade graphite are achieving reproducibility, and a graphite that will hold up under irradiation. On the basis of work done to date it appears that fabrication of the fuel element matrix by binding of graphite flours will result in a product of maximum reproducibility. From the result of irradiation studies made by Battelle, it has been concluded that experimental graphites made from a special laboratory petroleum coke, a sugar carbon or a phenolic resin carbon, showed the best stability under low temperature irradiation.

1.1.2 Fissile and Fertile Material

Reactors using graphite-uranium fuel elements that are under construction or planned have the fissile material in the form of U_3O_8, UO_2 and UC_2. The choice of the form in which the fissile material will be used is dependent, to a great extent, upon the anticipated operating temperature.

Temperatures at which the reactions occur, converting one form of the oxide to another and to the carbide are variously reported. What is usually lacking is a definition of particle size, which dictates the rate at which the reaction at a particular temperature will occur. The reactions and temperatures quoted here are as reported by Johnson, Falkerson and Taylor (11), unless otherwise noted.

Liquid impregnated elements are usually made with uranyl nitrate hexahydrate, $UO_2 (NO_3)_2 \cdot 6H_2O$. Uranyl nitrate is decomposed by heat to UO_3 at 225°C. This powder is then reduced at 800°C to UO_2. The fine grain oxide which results generally has a particle size of less than one μ. According to Mellor (12) the uranyl nitrate hexahydrate melts at 60.2°C; gives up two molecules of water of crystalization at 100°C; 2 more at 118°C and the last two at 170°C. The nitrate radical breaks down at 225°C, leaving the orange salt, UO_3. X-ray defraction studies at Argonne (3) indicate that heating at 800°C causes the oxide to take the form of U_3O_8, which on continued heating goes to UO_2.

According to Johnson et al (11) U_3O_7 is formed by a $UO_2 + O_2$ reaction at 200°C. Upon continued heating in the presence of oxygen, U_3O_8 is formed at 400°C. This is stable until approximately 1200°C at which point the U_3O_8 reverts to UO_2. It is generally conceded that U_3O_8 should not be used above about 500°C, the exact temperature being dependent upon particle size.

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UO_2 is available in many particle sizes ranging from submicron to several hundred microns. It can be fabricated in bulk shapes ranging from tiny pellets to rods an inch in diameter and a foot long. The forms of interest in dispersed fuel elements are "ceramic" and "high fired" grade, the former being a powder and the latter a nodule. The "high-fired" grade is preferred by some (10) in the fabrication of admixtured elements due to its superior blending qualities.

UO_2 converts to UC and UC_2 in the presence of carbon at 1750 to 1800°C. If the fuel is to be in the form of UO_2 it is necessary that temperatures of the order of 1200 to 1400°C not be exceeded in the fabrication step. In order to produce an admixtured element having fissile material in the oxide form, it is mandatory that the matrix be prepared by mixing graphite flour, UO_2 and a binder and baked at a temperature below 1200° - 1400°C which will carbonize, but not graphitize the binder.

If fuel is to be in the form of the carbide, it can be added as such, or added as an oxide, and then converted during graphitization of the fuel element. The UO_2 will convert to a mixture of UC and UC_2 at between 1750 and 1800°C. In general, UO_2 is preferred for the initial steps since the necessity of protecting the uranium carbide against conversion to the oxide or nitride is avoided. UC and UC_2 melt at about 2300 to 2400°C. In selecting a fuel for his high temperature reactor Daniels (13) chose the carbide form "because it can be heated to temperatures above 3600°F (2000°C) in contact with graphite and helium without appreciable reaction, vaporization or formation of gaseous products. The carbide oxidizes easily in air above 500°C to yield an oxide which dissolves readily in nitric acid to give uranyl nitrate without evolving gas."

Uranium carbide has not received too much attention as a fuel material, principally because few, if any, reactor concepts involved the temperatures that made the use of the carbide necessary. The objection to the carbide has been its instability. BMI find that UO_2 going to UC + UC_2 in situ does not introduce an objectionable gassing problem. They also find that UC + UC_2 in graphite is much more stable than the carbide alone. They have exposed carbide-graphite samples in air for six months with no change in composition where UC_2 would have converted to UO_2 within this period. However, work at BMI has led them to conclude that uranium losses from a carbide element would be of the order of 30 to 50 times greater than losses from an oxide element, pointing to the desirability of a protective coating on a carbide element.

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Degussa have prepared and irradiated fuel elements for the BBC-Krupp reactor using uranium carbide, which is reported as being the monocarbide. No information is available regarding the material or the results of the irradiation work.

1.1.3 Coatings

Coatings are of primary interest for the fixed graphite parts of the PBR. Graphite oxidizes in the presence of high temperature steam and is therefore susceptible to damage in a system when steam could be released to the reactor as the result of an accident. The water gas reaction rate is a function of temperature and in addition, the effect of radiation is not known. Therefore coatings will be used on those fixed graphite parts which would be difficult if not impossible to replace in event of steam damage.

If coatings can be developed for fuel elements which would reduce fission product leakage by several orders of magnitude, or to the point where activity release in the primary system is inconsequential, then their use should be considered for fuel and blanket elements. This is, however, an economic consideration, taking into account the neutronics, containment, maintenance, fabrication and reprocessing.

Several coatings for the protection of graphite and/or the retention of fission products are being developed by different manufacturers. Results to date appear promising but the work has not been carried to the point where it is conclusive.

Probably the best known of such materials is silicon carbide which oxidizes slowly in air at about 3400°F, increasing rapidly as the temperature is increased to 4000°F at which point it decomposes. Due to the difference between the coefficient of expansion of silicon carbide and graphite it has been difficult to apply coatings which would not crack although progress in the solution of this difficulty is reported by the Bureau of Mines (14).

Silicon carbide increases in weight upon oxidation in steam. BMI (15) report this weight increase to be of the order of 0.15% in 24 hours with steam at 1000°C. The significance of this weight increase with respect to PBR operation is not known at this time.

Minnesota Mining & Manufacturing Company have under development a coating identified by them as 3M "Ceramic S" which we believe

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to be a siliconized silicon carbide. Tests performed and reported by them (16) on coated graphite sample are as follows:

Reagent	Time-Hours	Results
96%-H_2SO_4	100	no weight loss
96%-H_2SO_4+ 2% HNO_3	100	no weight loss
Aqua Regia	100	no weight loss
900°C Steam	100	no weight loss
900°C CO_2	150	no weight loss

Of these tests the one of primary interest is 100 hours exposure to 900°C steam with no measurable weight loss. If these results hold for production coatings and if such coatings can be obtained economically, it would eliminate any concern over oxidation of graphite in event of a steam leak into the system.

The Engineering Science Division of American Metal Products Company of Ann Arbor, Michigan are developing a "diffusion resistant coating" for graphite parts (17). It is reported that they are able to diffuse pure carbides of hafnium, molybdenum, titanium and zirconium into the surface of a graphite part to a controlled depth and distribution. All of these coatings are good to 1000°C in air. It has been found that two coated pieces will adhere to each other at 2600°C. They can withstand a thermal shock test wherein they are heated and cooled in a ten minute cycle between 600 to 2800°C. Metal loadings equivalent to 40 mg/cm^2 are required to produce a diffusion resistant coating. The effects of this loading on nuclear characteristics remain to be evaluated. They advise us that they are making coated graphite elements for the Bureau of Mines test program.

BMI have not done any extensive work on coatings as such but have recognized the problems of retaining fuel and fission products. Their first interest was retaining fuel at temperatures of the order of 3000°F. Based on some preliminary work, which showed that a loss of fuel occurred at an insignificantly slow rate, they concluded: a) fuel loss was further reduced by placement of uranium compounds in a relatively massive form, well below the surface of the element; b) that surface coatings could decrease fuel loss by a factor of 10 or more at 3000°F and c) fuel losses from UC_2 loadings were 30 to 50 times those from UO_2 loadings (18).

One of their first coating investigations was reported in (19) where a Cr_3C_2 coated graphite tube axially loaded with a UO_2 rod showed no detectable uranium loss in six hours at 2500°F in argon. Subsequent reports (20) indicated a loss of $0.5 \mu g/cm^2/hr$ in 25 hours and no loss in the next 75 hours. An uncoated graphite tube containing a UC_2 rod showed losses of about ten times this value in the first 25 hours. Further work (21) with a UO_2 rod sheathed in graphite and exposed in a flowing nitrogen stream at 2000°F showed a loss of $0.54 \mu g/cm^2/hr$ in the first 24 hour period. Correlating these data indicates that either the losses were within the resolution of the measuring equipment or that the Cr_3C_2 coating was of no value.

This work was stopped in June, 1957 due to a change in emphasis in the contract. Present activity at BMI in this field concerns alumina urania fuel mixtures in graphite (22) (23). This work is based on the argument that urania clad with alumina appears to be superior to urania graphite with respect to fission product retention and at the same time, radiation damage to the graphite is reduced. Incorporation of discrete UO_2 particles in a matrix of dense Al_2O_3 and subsequent dispersion in graphite would take advantage of these characteristics of alumina while maintaining the mechanical characteristics of graphite. In summary, alumina-urania fuel mixtures in graphite offer the possibility of a)retaining fission products, b) preventing recoil fission products from causing radiation damage in graphite and c)eliminating cladding.

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1.2 Manufacturing Methods

1.2.1 Impregnation

The proposal to incorporate uranium in a graphite matrix by impregnation can be found in one of the earliest disclosures made of a power reactor (1). Following this proposal the Argonne National Laboratory investigated the fabrication of fuel elements by this method. Lack of high temperature graphitizing facilities at ANL and the relative ease with which preformed graphite pieces could be fueled by impregnation led to an extensive experimental program along these lines which was carried out by the Chemistry Division of ANL. Shortly thereafter similar work was done at the Aerophysics Laboratory of North American Aviation. More recently, interest in this method of graphite fueling has developed at Los Alamos and at Livermore. Since all the steps in an impregnation operation are easily adapted to remote processing techniques, it seems reasonable at the present time to re-examine this process in connection with the use of recycled fuel of low decontamination factors.

Early Argonne work (2) developed a process for impregnating the pores of graphite with a solution of a uranium salt which could then be converted to a form stable at reactor conditions. Uranyl nitrate was chosen because of its high solubility and the fact that it is readily converted to uranium oxide which is stable at reactor conditions. The process consisted of the following steps:

1. Machine graphite to desired shape.
2. Boil graphite in water to remove surface dust introduced during machining.
3. Heat in helium atmosphere at 800°C to remove interstitial and adsorbed water.
4. Evacuate sample and soak in aqueous uranyl nitrate solution.
5. Dry slowly under equilibrium conditions.
6. Fire at 800°C to convert uranium first to U_3O_8 and then to UO_2.

A modification of the process at step 4 consists of soaking the graphite in a refluxing solution of uranyl nitrate in order to eliminate the evacuation step. In general it was found that when aqueous solutions (1 molar) were used, only about 65% of the voids were filled as contrasted with about 90% of the voids being filled during evacuation impregnation. This condition

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was found to hold for several different types of graphite. It was also determined that reflux impregnation resulted in greater variability from piece to piece than did evacuation impregnation. It was recognized that reproducibility of results depends chiefly on graphite density and pore structure characteristics, i.e. presence of interconnected pores, etc., however no explanation has ever been presented for the difference in reproducibility in the two methods of impregnation. An indication of the effect of graphite density and the degree of reproducibility can be seen in the data of Figure 1-2.

Figure 1-2
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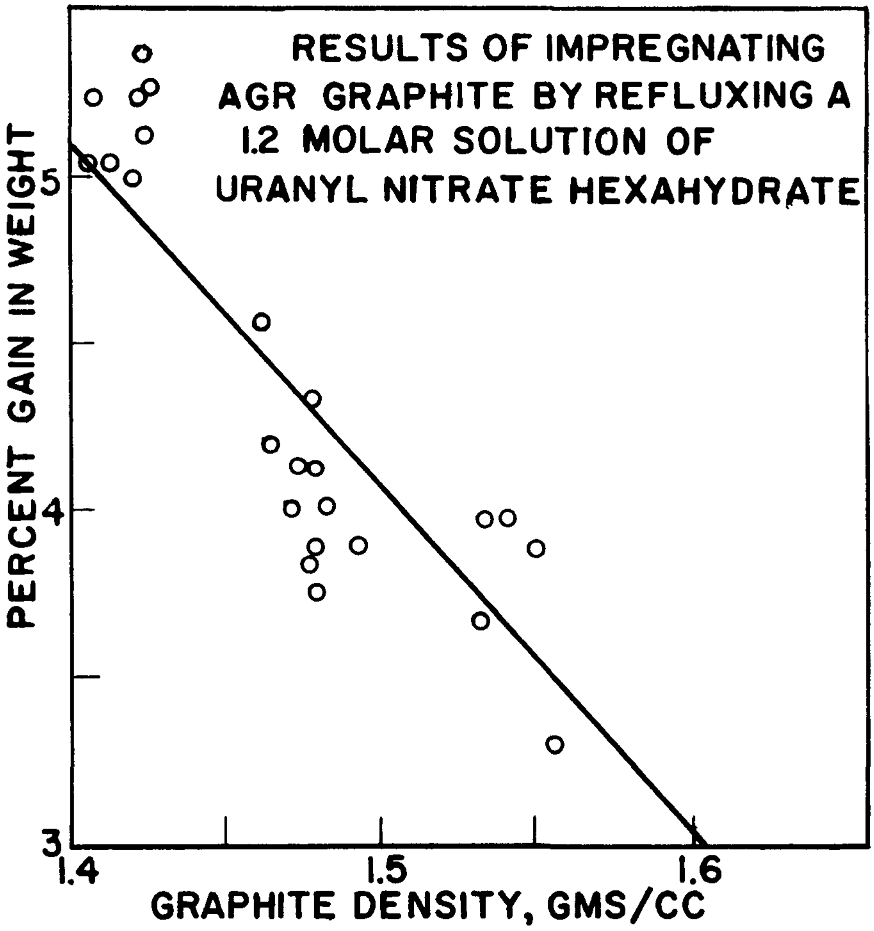

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ANL was concerned over the uniformity of the distribution of uranium within the graphite and concluded that the critical step was the drying operation. They report that essentially uniform distribution was achieved when slow drying was used and that the uranium concentrated in a skin near the surface when rapid drying was used.

North American Aviation also experimented with the impregnation of graphite with uranium. Apparently NAA had much more trouble than ANL in obtaining uniform distribution of the uranium in graphite (25) (26). When impregnating with aqueous solutions, they found surface concentrations to be about 8 times the average concentration and concluded that the uranium migration occurred during firing at 800°C. It was proposed to alleviate this problem by precipitation of the uranium as ammonium diuranate before drying the sample. This was done by soaking the impregnated part in ammonia gas for 24 hours prior to drying. The following reactions indicate the process:

$$2\,UO_2(NO_3)_2 + 6H_2O + 6NH_3 \longrightarrow (NH_4)_2U_2O_7 + NH_4NO_3 + 9H_2O$$

$$(NH_4)_2U_2O_7 \xrightarrow{150^\circ C} 2UO_3 + 2\,NH_3 + 3H_2O$$

$$3UO_3 \xrightarrow{800^\circ C} U_3O_8 + 1/2\,O_2$$

This apparently reduced the ratio of surface to average concentration from 8 to 2. It would seem that careful control of the drying operation does more good and obviates the need for this additional step.

One of the early impregnation procedures (27) developed by NAA resulted in uniform impregnation, however the procedure was not amenable to production of any large number of fuel elements since it involved sealing the graphite and uranyl nitrate hexahydrate crystals in an evacuated glass tube, firing at 800°C to convert the uranium to UO_2, followed by breaking the sealed glass capsule and machining off the excess UO_2.

The most uniform distribution of uranium resulting from impregnation and reported to be as uniform as the porosity of the graphite will permit, has been reported by NAA (28). This was achieved by impregnating graphite with uranyl nitrate dihydrate in tertiary butyl alcohol, quick freezing of the impregnated block in liquid nitrogen, sublimation of the solvent below the melting point of the solution by a "freeze dry" process followed by heating at 725°C to convert to UO_2.

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More recently LASL (7) has done work on the impregnation technique and has prepared graphite uranium fuel elements varying from 13 mm dia. to 1-1/2" dia. using a process which is only a minor modification of the ANL process described above. LASL reports a reasonably uniform distribution of the uranium and has stated in a personal communication that this is quite dependent on the detailed conditions used during the drying step.

Uniformity of fuel distribution is not as important in the PBR element as it would be, for instance, in a uranium graphite block having numerous coolant holes. Actually, a spherical element having a fuel concentration that varied proportionately with radius would operate at a lower central temperature than an element with a uniform fuel distribution.

Los Alamos Scientific Laboratory has impregnated graphite spheres for Sanderson & Porter as shown in Figure 1-3 with the results indicated in Table 1-3. LASL reported that samples AUC-1, 2, 3, & 4 were not fully submerged during impregnation due to an equipment fault and therefore the variations in UO_2 loading are not indicative of what might be expected during production operations. The molded samples M-1, 2 & 3 were thought to have a dense skin resulting from the molding operation and it was therefore expected that they would resist impregnation. National Carbon Co. who had furnished the molded samples for other purposes, reported that this was to be expected since the molded graphite was deliberately made to have a minimum number of interconnected pores since it was being used in the fabrication of elements by admixture. The results are therefore not to be interpreted as a skin effect but rather are characteristic of the type of graphite used. The remaining results show a range of ± 10% around the mean which is a smaller variation than indicated by ANL experiments.

The Engineering Sciences Division of American Metals Products Company of Ann Arbor, Michigan are developing impregnated fuel elements by the gaseous diffusion of UF_6 into a graphite body with subsequent conversion to the uranium carbide (17). This work is in its early stage and no information regarding the mechanical characteristics of the finished product is available. By proper production technique it is reported that the distribution of UC can be controlled within the final body. Due to the fact that the uranium is in the form of the carbide it is necessary to apply a diffusion coating, as discussed in Section 1.1.3, to prevent the conversion of the carbide to other forms.

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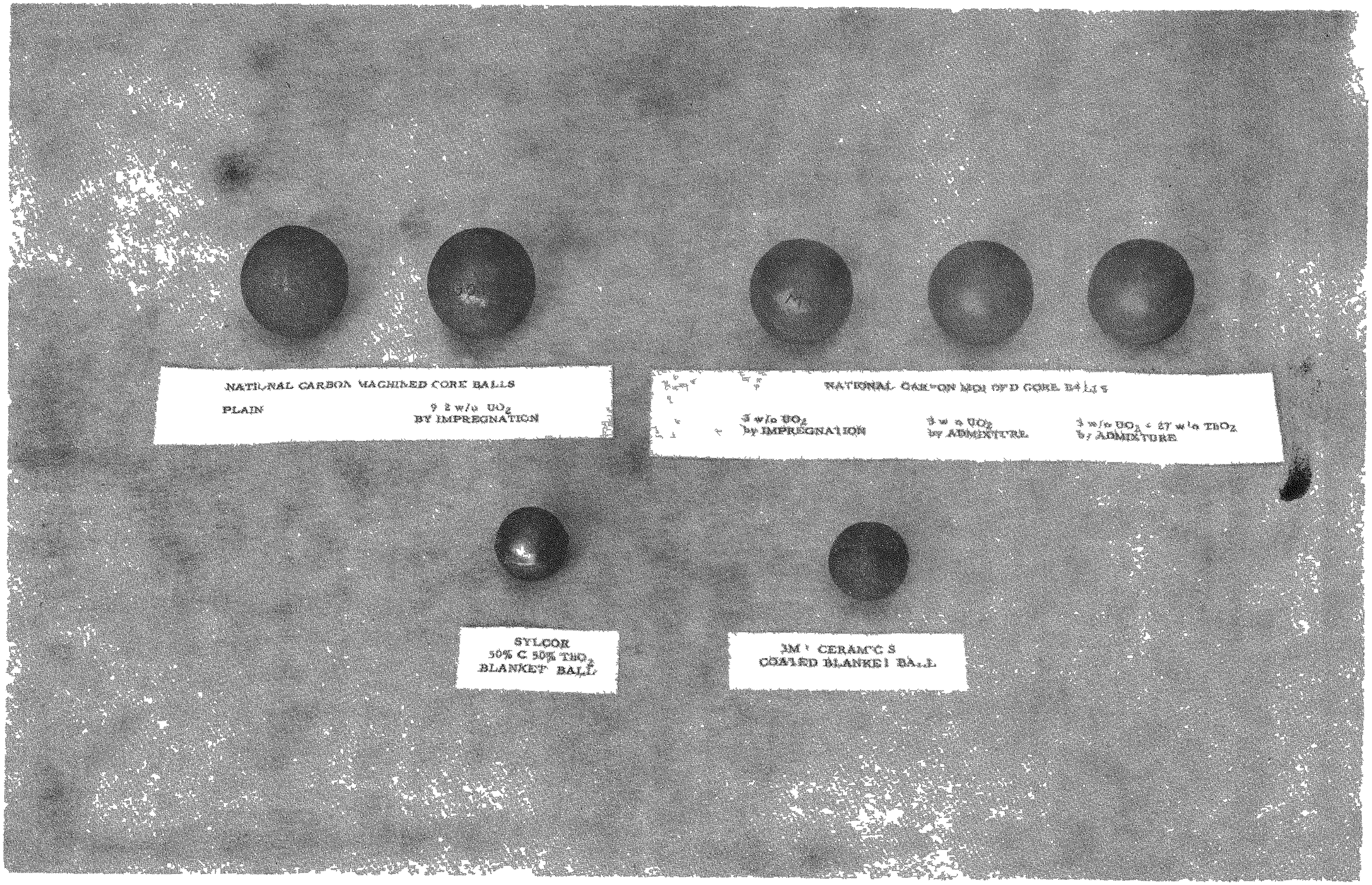
NATIONAL CARBON MACHINED CORE BALLS
PLAIN
9.2 w/o UO_2 BY IMPREGNATION
NATIONAL CARBON MOLDED CORE BALLS
3 w/o UO_2 by IMPREGNATION
3 w/o UO_2 by ADMIXTURE
3 w/o UO_2 + 27 w/o ThO_2 by ADMIXTURE
SYLCOR 50% C 50% ThO_2 BLANKET BALL
3M CERAMICS COATED BLANKET BALL

Figure 1-3 UNCLASSIFIED

TABLE 1-3

Results of Graphite Uranium Impregnations at LASL

Type & Sample No.	Mfg. Method	Density gr/cm^3	Wt % UO_2 1st. impregnation	Radiographic Analysis
AGOT-1	Machined	1.70	10.6	
AGOT-2	"	"	10.0	edge-171 mg/cm^3 center-140 mg/cm^3
AGOT-3	"	"	9.2	
AGOT-4	"	"	9.5	
AGOT-5	"	"	10.5 (12.3)*	
AUC-1	"	1.74	3.2	
AUC-2	"	"	4.0 (6.1)*	
AUC-3	"	"	5.5	
AUC-4	"	"	7.0	
AUC-5	"	"	11.4	
AGOT-M-1	Molded	1.66	0.55	
AGOT-M-2	"	"	3.00	
AGOT-M-3	"	"	2.90	edge-95 mg/cm^3 center-near zero

* Total after 2nd impregnation.

1.2.2 Admixtures

Experimental work on the fabrication of fuel elements by admixture was initiated at Battelle Memorial Institute in 1950. The greater part of their work has been on elements fabricated with the use of plastic resin binders although work has been done with pitch binders. They have prepared and irradiated bodies containing both UO_2 and UC_2 by both methods.

The process developed by Battelle was first reported in (29). It consisted of preparing a mix of graphite flour, UO_2 and phenol formaldehyde binder, molding under approximately 20,000 psi pressure and baking at about 1400°C to carbonize the binder. The advantage of this process lies in the use of a plastic resin as a binder which permits the forming of the mixture at room temperature and obviates the necessity of supporting the molded body during the subsequent firing step.

The Battelle work is quite extensive and includes a study of UO_2 particle size by incorporating the material by impregnation, as UO_2 powder which is milled and sieved to select different size agglomerates, and high-fired UO_2 where the particles are spherical in shape. Artificial graphite flour of several types has been used as the matrix material and binders used have been the phenol formaldehyde resins as well as pitch. Shapes have been extruded and molded and final baking has been done at various temperatures in order to either carbonize or graphitize the binder.

Their work has indicated that the flexural strength or modulus of rupture is of about the same order of magnitude as reported in Table 1-2. References (30) and (31) discuss results of a program designed to improve graphite as a reactor material by varying the raw materials and fabrication procedures and as a result of this work Battelle are of the opinion that the physical properties can be tailored to fit specific requirements with a fair degree of reproducibility.

BMI have fabricated and irradiated graphite-uranium samples of their own manufacture using reactor grade graphite flour as filler and BV 1600 bakelite resin as a binder, loaded with UO_2 and carbonized. They have also fabricated and irradiated samples made with petroleum coke, pitch binder and UO_2 where the samples were graphitized and therefore the uranium was finally in the form of UC_2. These samples were irradiated at the MTR and returned to BMI for examination.

For the past few years the National Carbon Company have been active in a research and development program on the fabrication of urania and/or thoria bearing graphite bodies by the admixture method. A wide variety of shapes have been produced such as blocks, cylinders and spheres and materials containing from 3 to 60 weight percent of oxides have been produced. Figure 1-3 shows two 1-1/2" nominal diameter spheres produced in connection with the PBR study containing in one instance 3 weight percent of UO_2 and in the other 3 weight percent of UO_2 plus 27 weight percent of ThO_2.

These samples were prepared by hot molding a mix of AGOT grade graphite flour, a high-fired oxide and pitch binder and then baking to carbonize the binder. Table 1-4 presents comparative properties of similar samples with different weight percents of UO_2 at two final baking temperatures. In those samples baked at 1450°C the binder is in the form of carbon and the uranium as an oxide. In those samples baked at 2800°C the binder is graphitized and the uranium is in the form of a carbide.

The Sylcor Corporation have been studying the preparation of graphite-uranium fuel elements under AEC contract for the past year. They have fabricated experimental quantities of blanket and core balls for the 125 eMW-PBR by admixture. In general, their process has been to mold a mix of No. 38 Atcheson graphite flour, thoria and/or urania, and a bakelite resin binder. The mix is baked at about 100°C to set the binder and then at about 750°C to carbonize the binder. One inch blanket balls, containing 50 weight percent of ThO_2, are shown in Figure 1.4 together with 100 X and 250 X photomicrographs. These balls have a density of 2.802 to 2.823 grams per cc.

Los Alamos Scientific Laboratory have also been developing a "room temperature" method of preparing a uranium-graphite mix for subsequent firing. Their procedure consists of mixing 85 parts of graphite flour (Great Lakes 1008) with 15 parts of lamp black, plus the required amount of high-fired UO_2, with Plaspreg (Furane Plastics product). This is all done in a dry box and results in a mass of the consistancy of wet sand. It is removed from the dry box, pressed at 6000 psi

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TABLE 1-4

Properties of Molded Uranium-Graphite Bodies

Baking Temperature - 1450°C - Carbonized Binder

Weight % U	Bulk Density gr/cm^3	Thermal Conductivity BTU/Hr-Ft-°F	Modulus of Rupture psi	Compressive Strength psi
0	1.72	12.7	6600	15,800
3.8	1.75	11.5	5470	18,600
11.7	1.88	9.8	5070	18,990
19.9	1.93	10.4	4290	14,100

Baking Temperature -2800°C - Graphitized Binder

Weight % U	Bulk Density gr/cm^3	Thermal Conductivity BTU/Hr-Ft-°F	Modulus of Rupture psi		Compressive Strength psi
			w	a	
0	1.70	26.0	6000	4680	10,070
3.9	1.78	26.0	5140	3800	9,060
11.7	1.89	32.0	4470	3400	8,300
20.8	1.99	33.0	3440	2060	5,570
23.7	2.09	37.0	-	2280	5,520
30.3	2.22	-	4230	-	5,940

w - with grain a - against grain

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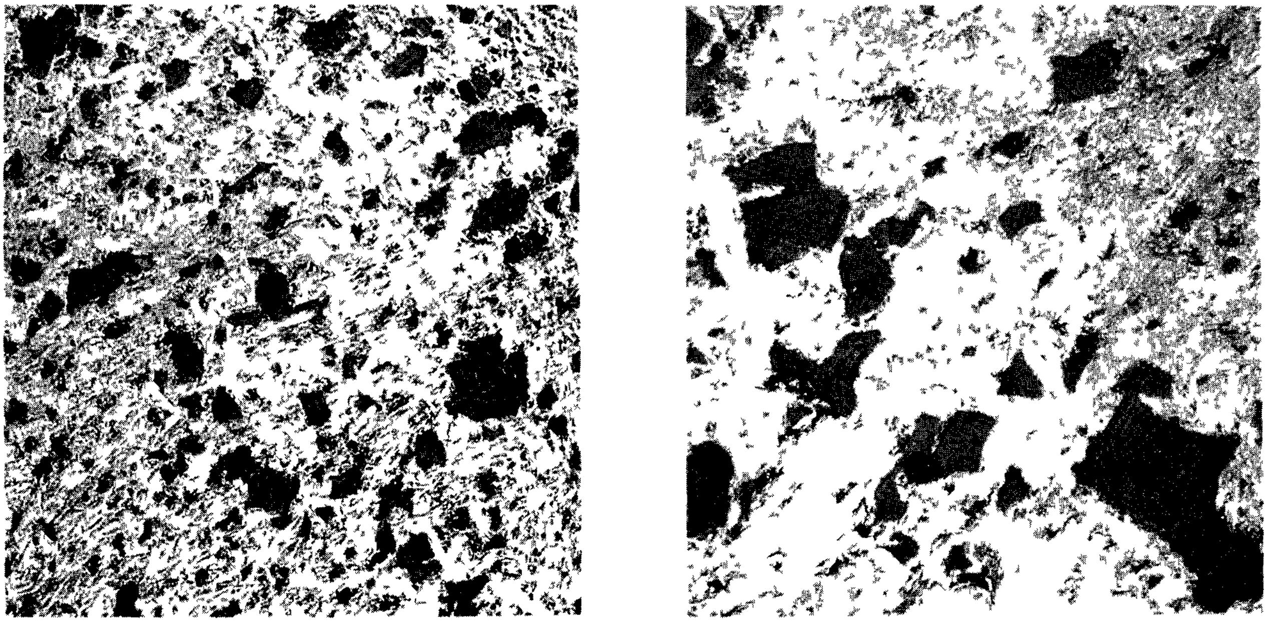
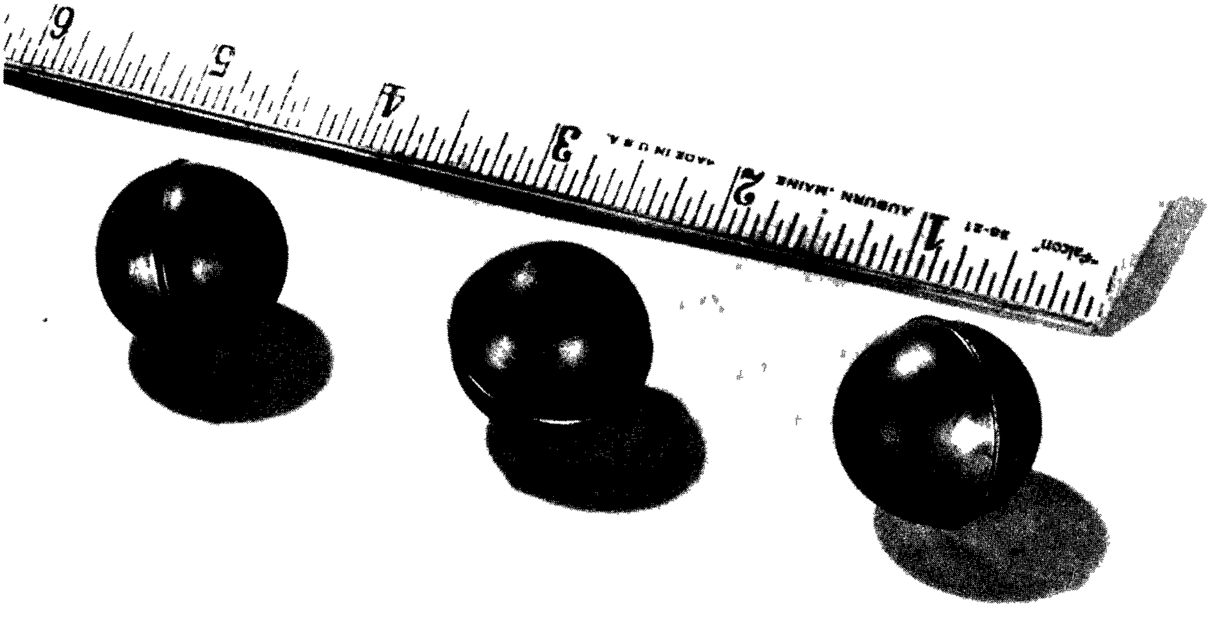
Figure 1-4 UNCLASSIFIED



Photomicrograph of Thoria-loaded Graphite

100 x

250 x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5
4
3
2
1
MADE IN U.S.A.
AUBURN, MAINE
Falcon

Fueled Graphite Balls Fabricated by Sylcor.
Balls Are Approximately 1" Diameter and Contain
50 w/o ThO_2 in Graphite

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and fired to carbonize the binder. The resulting graphite has a density of approximately 1.8 grams/cm^3. They find that Mallinckrodt "high fired" UO_2 is superior to the ceramic grade as the latter agglomerates while the "high-fired" grade, consisting of spherical particles, mixes well with the graphite flour. (10)

The Great Lakes Carbon Company are presently manufacturing the fuel elements for TREAT, a pulsed research reactor being built by ANL. These elements are uranium graphite fabricated by admixture. They contain 0.24 w/o of 93.1% enriched U-235 in the form of U_3O_8. Particle size is approximately 100 μ.

1.2.3 Lumping

Work on the fabrication of lumped fuel elements has not been as extensive as fabrication by impregnation or admixture. Dr. Farrington Daniels has done some work in this field reported in (13), where UC_2, graphite and stearic acid were pelletized and placed in a graphite tube which was plugged. Degussa have done similar work along these lines for BBC-Krupp who are fueling their reactor with lumped elements. These elements are made by drilling a hole in a 50 mm (2 inch) graphite ball, filling it with a UC_2 + C mix, plugging the hole, and heating the ball to graphitizing temperatures. Fuel elements of this type loaded with natural uranium have been irradiated at NRX (Chalk River, Canada) for seven weeks in a 5 x 10^{13} neutron flux. Boron was inserted in the elements in an attempt to further simulate fission fragment damage.

Minnesota Mining & Manufacturing are investigating several ways of fabricating lumped elements coated with their 3M "Ceramic S" coating, designed to reduce the escape of gaseous fission products. These methods are:

a) encase a pellet of UO_2 and/or ThO_2 in a graphite shell.
b) encase a pellet of graphite, UO_2 and/or ThO_2 in a graphite shell.
c) encase a pellet of graphite and ThO_2 in a UO_2 shell and then a graphite shell.
d) encase a pellet of UO_2 in a graphite-ThO_2 shell and then a graphite shell.

They have fabricated fully enriched UC_2 lumped fuel elements for irradiation in the MTR for a customer. These elements, coated with 3M "Ceramic S", were leaktight to helium at room temperature. They are in the process of fabricating about 100 UO_2 lumped fuel elements, both coated and uncoated, for study and irradiation by the Solid State Physics Group at Oak Ridge. Similar fuel elements have been made for other customers for irradiation study.

Battelle have done a modest amount of work on lumped fuel elements made by two methods: a UO_2-graphite matrix clad with graphite and a UO_2 sintered pellet clad with graphite. Spheres were of 5/8 inch diameter with a 0.4 inch diameter core. It was found that the UO_2 sintered pellet elements cracked while baking while those fabricated with the UO_2 graphite matrix did not crack. (32)

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1.3 Fuel Element Stability

The characteristics of graphite uranium fuel elements of principal interest in the PBR design are the impact strength and tendency to self-weld, which affect the loading and unloading of the reactor; and the crushing strength, tensile strength, thermal conductivity, surface stability, fuel retention and fission product leakage, which affect reactor operation. Of prime importance is the effect of irradiation on these characteristics.

The majority of these characteristics have been investigated for various uranium-graphite systems at one time or another. Post-irradiation effects, as a function of temperature and exposure, are either scattered or completely lacking. The substance of this past work is such that although final engineering design could not be performed with confidence based on these data, it does however, prove the validity of the fuel concept and provide a foundation for specific experimental work.

1.3.1 Impact Strength

The first question of fuel element stability concerns impact strength, as the PBR is designed to be loaded and unloaded by gravity. As reported in Section 3.0 of Part I of this report, we have run simple drop tests on machined and molded graphite balls both with and without UO_2. Results indicate that free falls ranging from 98 to 238 feet are possible without fracture, depending upon the method of manufacture, fuel loading and whether the ultimate fracture resulted from a drop with or against the grain.

1.3.2 Self-welding

There has been some concern expressed over the possibility of adjacent fuel balls fusing together or "self-welding" to each other. It has been suggested that this could come about because of the temperature and pressure to which the balls will be subjected in the core, or through the possibility that uranium or thorium might migrate to the surface and thereby provide the adhesive agent. It is highly unlikely that the balls would self-weld through a combination of temperature and pressure that would exist in the PBR, as temperatures of 4000°C and pressures of 50,000 psi are required to weld graphite under controlled conditions. It is equally unlikely that uranium would migrate to the surface in sufficient quantity or form as to result in "self-welding" of fuel balls.

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There is some concern over the possibility that a carbon transport phenomenon could provide a mechanism to promote fusing of adjacent balls. This seems unlikely since the points of contact will operate at higher temperatures than surrounding areas and would therefore be expected to lose rather than gain carbon if mass transport did occur.

1.3.3 Fuel Loss

There is some evidence to indicate that there might be a uranium loss from the surface of a fuel element, assuming the surface contained fissile material, i.e. there was no cladding or non-fuel bearing coating on the element.

A number of experiments have been performed to determine the loss of uranium from uranium-graphite specimens, much of which has been at temperatures higher than those contemplated for the PBR. ANL (34) reported some work in 1948 which was done in the temperature range of 1500 to 2600°F on graphite cylinders of 3/4 inch diameter by 1-1/2 inches long and of 1.37 to 1.43 density impregnated with U_3O_8. These cylinders were induction heated and subject to a flowing helium stream and the uranium loss determined from filter analysis. Examination of the results indicates that the measured loss is probably within the resolution of the data and varies from .00086 to .0026 $\mu g/cm^2/hr$ at 1500 to 2000°F sample temperature.

This work showed that the amount of uranium loss was strongly dependent upon the disposition of the fuel in the graphite. At the time the work was done, the impregnation process had not been developed to a point where uniform impregnations were being obtained which accounts for scattering of the initial data. It was only after the outer layer containing the heavy uranium loading was machined off that reasonably uniform loss data was obtained. It was also concluded that trace oxygen in the helium resulted in increased vaporization rates, due probably to a change in form of the uranium oxide and loss of surface carbon, thereby leaving uranium exposed.

BMI have conducted a variety of experiments on this subject. No loss of uranium was observed during 2 hour graphitization treatments (4000° - 4500°F) of admixtured specimens using UO_2 powder but radiographs did reveal significant coalescence of smaller particles into larger clumps (6). Varying uranium losses are reported (20) (21) (33) for UO_2 and UC_2 rods co-extruded in unfueled graphite subjected to flowing nitrogen or argon at temperatures of 2000°F to 2500°F. In the case of the

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UC_2 specimen, uranium losses dropped from 6.2 to 0.5 $\mu g/cm^2/hr.$ in a 100 hour test. In the case of the UO_2 specimen, initial uranium losses of 0.5 $\mu g/cm^2/hr.$ dropped to essentially zero in a period of about 48 hours. No significant difference was noted in the latter case when the specimen was coated with Cr_3C_2. It is not clear why the BMI data gave higher losses than the ANL data which was obtained under conditions seemingly more favorable for uranium loss.

1.3.4 Thermal Stress

Fuel elements in the PBR have been sized on the basis of thermal stress, using as design criteria presently accepted data for tensile strength, thermal conductivity, etc. National Carbon, among others, are doing some work on thermal stress and shock of graphite and have found it to be virtually impossible to rupture a graphite part by thermal stressing alone since slowly induced thermal stresses are relieved by plastic deformation. However, rapidly induced thermal stresses, i.e. thermal shock, can cause failure.

The Jet Propulsion Laboratory of the California Institute of Technology studied the heat flux-rupture limits of several graphites with internal heat generation up to surface temperatures of 5000°F, i.e. to graphitizing temperature (35). They concluded, based on a simplified calculational model, that the thermal stress in the specimens at failure was only half the known high temperature tensile strength of the same graphite and acknowledge that the infinite flat plate model used in computing the thermal stress did not adequately describe the stresses in a flat plate with edge cooling. This fact has been borne out by an extension of this investigation by LASL who suggest designing to 1-1/2 times high temperature tensile strength on the basis of work they have done.

The significance of this work in the PBR design is that if the graphite fuel elements plastically deform under thermal stress, then the tensile strength limitation can be removed and fuel elements can be sized on the basis of economics. While it would appear to be impossible

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to rupture a graphite sphere as the result of thermal stress, a knowledge of thermal conductivity in the pre- and post-irradiation period is desirable particularly with reference to the temperature gradient through the sphere and the resultant core temperature. Work that has been done on the change of thermal conductivity with irradiation is discussed later in this section.

1.3.5 Fuel Loading

The 125 eMW PBR has been designed with a limitation of 10 w/o of fissile and fertile material in the fuel elements and 50 w/o of fertile material in the blanket elements. While there is no question as to our ability to load graphite with these amounts of material, as sample elements of these loadings have been made for the PBR, there is the question of the effect of these weight loadings on the strength of the element.

Battelle Memorial Institute have fabricated a series of fuel samples by admixture in which fuel quantities were varied from 1 to 50 v/o of oxide. They found no change in tensile strength up to 18 v/o loading, which is the equivalent of 55 w/o in 1.7 density graphite (36) (37).

NAA (38) investigated the short time tensile strength of impregnated graphite where the uranium had been converted from UO_2 to UC_2 by heating. National Carbon grade C-18 graphite impregnated with 0.234 g/cc and 0.424 g/cc of uranium showed an appreciable reduction in tensile strength with temperature as compared with an unimpregnated specimen. National Carbon grades ECA and AUF impregnated with 0.147 g/cc and .009 g/cc of uranium respectively showed no decrease in strength as compared with the unimpregnated specimen. The decrease in strength of the C-18 grade with the higher loadings was attributed to the lower density and larger pore size of the C-18 grade. This, however, is not a completely satisfactory answer because of the wide variation in loadings and the different impregnating methods used. This is a question that should be evaluated at some time in a fuel development program.

1.3.6 Surface Stability

One of the questions pertaining to the stability of graphite-uranium fuel elements is that of surface stability. The Bureau of Mines are doing experimental work for a high temperature, helium-cooled process heat reactor utilizing graphite-uranium fuel elements. For the past year experimental work has been done on a simulated reactor at atmospheric pressure and 2500°F gas temperature. In connection with this work helium

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velocities up to 1640 feet per second were found to have no erosive effect on graphite elements at 2500°F (14). In addition, North American Aviation conducted tests on the erosion of graphite with a high temperature, high velocity helium stream. Tests with helium flowing through a nozzle at a throat velocity of 9000 ft/sec. and at a temperature of 2000°C, impinging on a graphite specimen heated to 2000°C, disclosed that for about 24 hours the machined graphite surface was subject to an initial erosion process in which loosely held particles, smeared into the surface during machining, were blown free. Once this process was completed, no further erosion was apparent (39).

In view of the fact that we are designing for actual helium velocities through the core of the order of 60 feet per second, and maximum gas and surface temperatures are of the order of 2000°F, there is no reason to believe we will be troubled by erosion if the fuel elements are properly cleaned before being placed in the core, and the surface condition does not change during irradiation.

1.3.7 Graphite Permeability

In connection with the general problem of impregnation, fuel loss, fission product leakage and coatings, we are interested in the permeability of graphite. BMI have investigated the effect of graphite composition and treatment on permeability and found that a body fabricated of skeletal graphite, having a density of 2.1 g/cc had a permeability two orders of magnitude less than a body fabricated of asphalt coke, having a density of 1.3 g/cc. Two bodies were fabricated of Korite coke and standard pitch. One body, pitch impregnated five times, had a permeability 1/20 of the non-impregnated body. Korite coke is of interest because of its dimensional stability and small change in thermal conductivity under irradiation (22).

BMI have also investigated the effect of final bake temperature on the permeability of fueled graphite bodies containing 2.7 w/o of enriched UO_2. From this work they have concluded that a body fabricated from a 79L* resin coke with a 79L* resin binder in a ratio of 100 to 15 had the lowest permeability as measured with argon. A skeletal graphite filler bonded with BV 1600 ** resin was equal, i.e. a minimum detectable

* Ironsides Co., Columbus, Ohio

** Bakelite Bonding Varnish, now identified as BKS-2600 gas flow corresponding to 0.028 x 10^{-5} Darcy, when both were baked at 2000°F (40).

1.3.8 Irradiation History

The success of the pebble bed reactor rests upon the stability of the graphite-uranium fuel elements under irradiation. During the past twelve years there have been a great number of experimental irradiations of graphite specimens fueled with uranium. This work stemmed from a variety of programs having various objectives. The objectives of much of the work fall into two categories: (1) fission fragment damage effects, as shown by changes in thermal or electrical resistivity, and (2) fission product diffusion studies under post-irradiation heating.

The earliest reported irradiations were by the ANL Chemistry Division from 1947 to 1949. This work originated as part of the Clinton Power Pile program. Some work was also done at ORNL under this program during the same period. Work was continued by the ORNL Physics Division from 1949 to 1951 using equipment similar to the earlier ORNL work. A joint program with BMI at ORNL on the variation of UO_2 particle size was reported in 1954. North American Aviation reported on numerous irradiations at LASL, HW and MTR during 1950 to 1954 as part of their work on a homogeneous research reactor and a high temperature power reactor. BMI worked under the Civilian Applications program from 1955 to 1957 after which time this work was continued as part of the GCRE program.

Table 1-5 has been prepared in an attempt to consolidate the pertinent facts about certain of these irradiations. The design conditions for the 125 eMW PBR are included as item 1 in this table. The total exposure for each specimen has been given as fissions/cc which is a common measure of potential damage to the graphite matrix.

The basic questions pertaining to the effect of irradiation on PBR fuel elements are:

(a) Will the graphite matrix retain its structural strength and resist crushing forces during operation and impact forces during loading and unloading?

(b) Will the fuel element surface continue to resist the erosive forces of the coolant?

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TABLE 1-5

SUMMARY OF IRRADIATIONS OF URANIUM GRAPHITE FUEL SPECIMENS

Item	Reported by	Reference	Date Reported	Where Irradiated	Quant.	Type	U-235 mg/cc	Enrich.	Form	Size	Temp. of Irradiation	Expt.(1) Info.	Fissions/cc	Comments	Item
1	——	——	——	——	—	—	13.7	Th/U=11	UO_2	1 1/2" sphere	2400°F max.	——	4×10^{19}	Design Conditions for 125 eMW-PBR (100 day; batch loaded)	1
											600°F min.	——	5×10^{18}		
2	Kierstead, Nagy	ANL 4006, ANL 4185	1947	CP-3	23	impreg.	up to 25	nat. & 30%	U_3O_8	.4 cm d. .4 cm L.	low	a, b, c	up to $2x10^{11}$		2
3	Wohlberg	ANL 4427	1949	HW	6	impreg.	20 to 26	30%	U_3O_8	.156" d. 10" L.	100°C	b, d, f	up to $2x10^{17}$		3
4	Hunter	MonN 442 TID 66	1947 1949	X-10	1	impreg.	300	93.6	UO_2	2 cm d. 5 cm L.	775°C, 630°C	a	$7x10^{18}$		4
					1	impreg.	300	93.6	UO_2	2 cm d. 5 cm L.	630°C	a	$3.5x10^{18}$		
					1	admix.	300	93.6	UO_2	2 cm d. 5 cm L.	630°C	a	$5x10^{18}$		
5	Billington, Primak & Berggren	ORNL 576, 693 and 1095; TID 10042	1949 to 1951	X-10	?	?	?	?	?	?	high and low	a	?		5
6	Kernohan	ORNL 1722	1954	X-10	12	admix.	90	93.6	UO_2	1/4"-5/16"-3"	30° to 88°C	a, b, c, e, f	$2x10^{17}$	Average UO_2 particle size: 586, 334, 94, 20	6
7	Smith and Young	NAA-SR-72	1951	LASL	1	impreg.	2.1	nat.	UO_2	——	low	g	$1x10^{12}$		7
8	Cubicciotti	NAA-SR-194	1952	LASL	many	impreg.	3.7	nat.	UC_2	.84 cm d. .064 to .52 cm t.	low	g	up to $1x10^{15}$		8
9	Doyle	NAA-SR-255	1953	LASL	many	impreg.	1.4	nat.	UC_2	5/8" Dia. -1/10" t.	low	g	$2x10^{14}$		9
10	Young & Smith	NAA-SR-232	1953	LASL	1	impreg.	.7	nat.	UO_2	——	low	g	$1.5x10^{12}$		10
11	McCarty and Steele	NAA-SR-223	1953	HW	2	impreg.	3.3	93.6	UO_2	1.3" d. x 4" L.	30°C	h	$8.4x10^{17}$		11
12	Hetrick, McCarty and Steele	NAA-SR-217	1953	HW	2	impreg.	24	93.6	UO_2	1 5/16" d. x 4 1/4" L.	25°-85°C surface 300°-350°C center	a	$7x10^{17}$		12
13	Durand, Klein, and Nykiel	NAA-SR-836	1954	MTR	1	impreg.	35	93.6	UO_2	1/2" d. x 3" 3" L.	1300°C	a, b, d, i	$1.1x10^{19}$		13
					1	impreg.	35	93.6	UO_2	1/2" d. x 3" 3" L.	1150°C	a, b, d, i	$4.1x10^{18}$		
					1	impreg.	35	93.6	UO_2	1/2" d. x 3" 3" L.	1000°C	a, b, d, i	$4.7x10^{18}$		
14	Moody, Taylor, and Johnson	ORNL 1778	1955	X-10	many	lumped	18	93.6	UO_2	1 cm d. x 0.1 cm.	low	g	($3x10^{17}$ nvt)	Many cladding types used, incl. SiC, TiC and ZrC	14
15	——	BMI 1181	1957	?	13	lumped	—	nat.	UC_2	——	low	g	($2.4x10^{15}$ nvt)	.03" dia. UO_2 rod coextruded with graphite	15
16	——	BMI	1958	MTR	1	admix.	40	93.6	UO_2	3/16x3/16x1"	1650°F	none to date	$1x10^{19}$		16
					1	admix.	40	93.6	UO_2	3/16x3/16x1"	1650°F	none to date	$5x10^{18}$		
17	Cockroft	Atomics and Nuclear Energy	1958	?	?	?	?	?	?	?	600°C	?	?	0.25% U burnup,	17
18	Krupp-BBC	——	1958	NRX	?	lumped	—	nat.	UC_2	5 cm sphere	low	?	($2x10^{20}$ nvt)		18

Note (1):
Key for Experimental Information
- a. Thermal conductivity
- b. Electrical resistivity
- c. Modulus of elasticity
- d. Dimensional change
- e. Weight change
- f. Post irradiation annealing
- g. Fission product diffusion (by post irradiation heating)
- h. Outgassing
- i. Crushing strength

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(c) What is the net effect on thermal stress within the fuel elements and will the decreased thermal conductivity affect stability by increasing the central temperature?

(d) How does radiation stability affect the choice between an impregnated element or the proper size fuel particles in an admixtured element?

(e) To what extent will uranium and fission products diffuse from the fuel elements?

Unfortunately, the results of experimental work to date do not yield conclusive answers to these questions. Only a few of the irradiations listed in Table 1-5 approach the exposure in the PBR. However, there are some results of interest. Regarding structural stability, it is worthy of note that none of the investigators reported the crumbling or disintegrating of the fueled specimens under irradiation. In only one instance was the change in crushing strength measured, Durand, et al. (Table 1-5, item 13) and in this case the crushing strength increased from about 7000 psi before irradiation to about 8500 psi after irradiation.

No reference to impact testing is made in any of the reports.

Since none of the specimens were irradiated in a high velocity gas stream, there is no information on erosion. However, Durand, et al. (Table 1-5, item 13) did observe a thin black powdery surface after irradiation which was unlike the shiny appearance of virgin AUF graphite. They postulated that this could have been due to air leakage through metal-to-ceramic joints in the capsule. However, since fuel distribution within the specimen is not reported, the likelihood of higher surface concentrations of fuel with resulting increased damage cannot be discounted. No other observations of surface condition are reported.

Many of the experiments reported the relative change in thermal conductivity of the specimens during irradiation. Results are reported as k_o/k (initial conductivity/conductivity) as a function of exposure. For impregnated fuel elements, Hetrick et al. (Table 1-5, item 12) reported k_o/k values ranging between 15 and 30 for low temperature irradiation, Hunter's extended irradiation (Table 1-5, item 4) at 775°C gave k_o/k of 39, while Durand et al. (Table 1-5, item 13) showed k_o/k values of 2.1 to 1.4 at higher temperatures. Thus, the tendency to anneal fission

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fragment damage at higher temperature is shown. Fission fragment damage can also be reduced by using larger fuel particle sizes in an admixtured element so that many of the fission fragments are absorbed in the fuel particle rather than the graphite matrix. Kernohan's work (Table 1-5, item 6) showed an increasing value of k_0/k from 1.09 to 1.50 as a function of decreasing particle size in admixtured specimens.

Thermal conductivity decrease can affect the thermal rupture and central temperature of the PBR fuel elements. However, the effect of irradiation on the other factors which affect thermal rupture (i.e. tensile strength, plastic deformation, etc.) remain to be evaluated before the significance of the larger thermal conductivity decreases for impregnated fuel elements can be assessed.

1.3.9 Fission Product Leakage

The basic PBR concept assumes the use of unclad graphite elements containing fissile and/or fertile material. Whether such a system can be operated successfully with unclad fuel elements, or to what extent cladding will be necessary, is not known and will have to be determined during the course of fuel element and reactor development. As a minimum, surface fuel can be leached from the element. A second step would be to apply a thin protective coating, such as achieved by an impregnation with subsequent carbonizing. A third step would be a separate layer of cladding applied in the form of a shell. What will ultimately be required can only be determined through a development program.

Unclad graphite-uranium fuel elements will permit the leakage of some fission products to the primary coolant stream. Table 1-5 has listed several experimental programs which were conducted in an attempt to evaluate leakage rates.

Unfortunately, all of this work has been based on low temperature irradiation followed by post-irradiation heating and does not reflect the effects of simultaneous irradiation, isotope decay, and temperature. Furthermore, this work was done on relatively thin specimens of graphite-uranium. Consequently, the work to date will not permit a quantitative analysis of possible fission product leakage from elements of the PBR type, although they have provided a considerable broadening of knowledge in this field.

A qualitative picture of the problem can be had by examining the fuel element on a microscopic basis. The spacing between carbon atoms in a graphite crystallite is reported to be of the order of 50 A ($5 \times 10^{-3}\mu$). Uranium oxide in impregnated graphite has an average particle size which has been variously reported as $.5\mu$ and 1μ. The average of fission fragments in uranium oxide is generally given as 5μ and in graphite as about 15μ. Thus it seems reasonable to expect that in fuel elements fabricated by impregnation, the bulk of the fission products will be found rather deep within the graphite crystal. Leakage of fission products from the fuel element must involve some or all of the following processes.

1) Diffusion through pores in the fuel element structure.
2) Diffusion along grain boundaries in the UO_2.
3) Diffusion through the UO_2 crystal.
4) Diffusion along grain boundaries in the graphite.
5) Diffusion through graphite crystal.

It is not improbable to consider that as many as three of these processes must take place in series before a fission product is capable of circulating in the primary coolant stream. For example, the fission fragment which imbeds itself deep within a graphite crystallite must diffuse through the crystal to a grain boundary, then along a grain boundary to a pore in the structure and finally through the pore to the surface of the fuel element before it can join the main circulating stream. It is generally conceded that pore diffusion is quite rapid compared with inter- or intra-crystalline diffusion.

Cubicciotti (Table 1-5, item 8) reports that after irradiating AUF graphite impregnated with 520 mg/cc of UC_2, 17% of the xenon was found in the uranium carbide and 83% was found in the graphite. Obviously therefore, the diffusion data reported by Cubicciotti is essentially that of xenon diffusing through graphite. Fuel elements made by the admixture method permit independent control of the fuel particle size. Use of 100μ particles, for example, would result in relatively fewer xenon particles in the graphite and Cubicciotti's results would probably not apply.

Doyle (Table 1-5, item 9) investigated fission product diffusion from a graphite sample impregnated with UO_2 by heating the pellets after short-lived precursors had decayed. The results of his studies are indicated in the table below.

Percent of Activity Remaining in Uranium-Graphite
Samples Heated 2-3 Weeks After Irradiation

Time	24 Hrs.		12 Hrs.
Element	1500°C	1700°C	1900°C
Cesium	4.5	1.4	-
Strontium	1	.07	.1
Barium	6.5	1.5	.07
Iodine	60	15	.7
Tellurium	57	8	1
Pr-rare earth group	79	17	5
Yttrium	100	55	44
Cerium	100	76	30
Ruthenium	100	100	80
Molybdenum	100	100	100
Zirconium	100	100	100

It is somewhat surprising to note that iodine which has a normal boiling point of 363°F, seems to diffuse at a much slower rate than does cesium, strontium, barium, or tellurium, all of which have much higher boiling points.

Young (41) investigated the diffusion products from small uranium impregnated graphite plates during actual creation of these fission elements and their precursors. This was done by heating the graphite for 20 hours in the range of 1050°C to 1950°C while bombarding it with deuterons from a cyclotron. (This irradiation is not listed in Table 1-5.)

When fission occurs at high temperatures, the diffusion characteristics of those members of the chain immediately formed in fission, and their successive daughters, can have a strong bearing on the "whereabouts" or position of the latter members of the chain. Taking the chain of mass 91, for example,

$$\text{Kr-91} \xrightarrow[\text{9s}]{} \text{Rb-91} \xrightarrow[\text{short}]{} \text{Sr-91} \xrightarrow[\text{9.7 Hr.}]{} \text{Y-91} \xrightarrow[\text{57 d}]{} \text{Zr-91}$$

Doyle found that yttrium did not diffuse when impregnated graphite pellets were heated to 1500°C for 24 hrs. after precursors had decayed; whereas Young found that only 6% of the yttrium remained in his sample which had been held at 1500°C during fission. Young's conclusions were as follows:

1. Elements which were unaffected by precursor diffusion were Sr-90, Mo-99, Ru-103, Ru-106, Cs-136, Cs-137 and Ba-140.
2. Elements for which diffusion was apparently influenced by precursor diffusion were Y-91, Ce-141 and I-131.
3. Elements which apparently diffused to a greater extent during irradiation than would be expected but whose increased diffusion could not be explained by precursor diffusion are Zr-95, Nb-95, Te-127, Te-129, Ce-144 and the praseodymium group.

Moody et al (Table 1-5, item 14) ran fission product leakage tests on a number of body compositions and concluded from the results of their work that claddings could reduce the volatile fission product leakage by a factor of the order of 10^4 to 10^5. Clad thicknesses investigated were of the order of 0.3 to 1 mm. This is the order of thickness of the 3M "Ceramic S" coating described in Section 1.2.3.

Results of other coating investigations are covered in Section 1.1.3. In summary, it is known that gaseous fission products will diffuse from the fuel elements but the quantity and species that will diffuse from the spherical elements of the PBR are not known nor can they be deduced from the work that has been done to date.

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1.4 Reprocessing

The graphite-uranium fuel element of the PBR is sufficiently different from conventional fuel elements so as to make unnecessary several of the processing steps of the present standard procedures. Our search of the project literature has not uncovered any references which deal specifically with reprocessing fuel of the graphite uranium type. Scattered, unrelated programs have been uncovered which shed light on one or another of the chemical steps which would be part of such a reprocessing scheme.

Two basically different reprocessing schemes, one resulting directly in uranyl nitrate and one in uranium hexafluoride are envisaged as being suitable for this fuel. The former consists of either incineration of the graphite followed by dissolution of the ash in nitric acid, or crushing of the fuel element followed by leaching with nitric acid to recover the uranium values; the latter consists of treatment with bromine trifluoride followed by vaporization. In the former the resulting uranyl nitrate solution would be subjected to a modified Thorex extraction operation in order to separate U, Th and fission products whereas in the latter case, simple distillation should suffice. The relative merits of these several alternates have not been determined to the point where a preference can be expressed. Furthermore, since the fuel element specifications have not been finalized, reprocessing specifications must be kept tentative.

The following paragraphs detail the extent to which work has been done which is applicable to this reprocessing scheme.

1.4.1 Incineration

It has been standard practice in laboratory studies of graphite impregnation to incinerate the graphite and weigh the ash as a means of determining the amount of fuel added. This incineration has taken place in air as well as in oxygen.

To our knowledge, no one has studied the incineration of uranium bearing graphite on a commercial scale. Thus the problems associated with dusting, fly ash recovery, uranium accountability, etc., have not as yet been evaluated.

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The dissolution of uranium oxide has also been studied extensively and is believed to present no serious problems. The optimum solvent concentrations, temperatures, etc., must be determined, which fall in the category of routine process development.

1.4.2 Crushing and Leaching

If it should turn out that the dusting and fly ash problems associated with incineration present serious problems with uneconomic solutions, then the separation of the uranium and graphite can be effected by crushing and leaching. This is a straightforward operation about which there is much general information. The specific information which should be obtained is the particle size to which the fuel element should be ground in order to optimize the solution operation and the liquid retention associated with the ground solid. This retention will determine the wash conditions required to recover the uranium values and will influence the concentration of the final recovery liquors. Countercurrent multistage leaching would be used to hold the quantity of dilute liquors to a minimum. In any event economic considerations will influence the selection of process conditions, once the liquid retention is determined.

1.4.3 Electrochemical Solution

ORNL (5) conducted experiments in 1948 in the electrochemical solution of uranium contained in graphite. The graphite-uranium piece was made the anode in a cell employing concentrated nitrate acid as the electrolyte. AC full wave rectification was used, resulting in the graphite disintegrating and the uranium going into solution as uranyl nitrate. This is an interesting operation, however it is not clear that it offers any technical or economic advantages over crushing and leaching. The countercurrent washing of the solid waste will be present in either case, thereby giving rise to the large columns of dilute liquors which will require concentration.

1.4.4 Halide Volatility

The previous discussion pertains to the recovery of uranium as uranyl nitrate which is then separated from fission products by Thorex extraction. An interesting alternate involves the recovery of uranium as uranium hexafluoride which can be separated from fission

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products by simple distillation. Levenson (42) reports that bromine trifluoride at about 110°C is capable of dissolving the uranium from UO_2, from U-Mo alloys, from U-Si alloys and from $Mo\,Si_2$ coated material. NAA reports (5) the dissolution of uranium from uranium bearing graphite. The simplicity of the subsequent fission product separation is apparent upon examination of the boiling points of several fission product fluorides.

SiF_4	65°C
TeF_4	38°C
$I\,F_4$	4.5°C
$Mo\,F_6$	35°C
$U\,F_6$	64°C
$I\,F_5$	97°C
$Nb\,F_5$, $Ru\,F_5$	> 200°C
Th, Zr, Cs, Sr, Ba, Rare earth fluorides	> 500°C

Thus most fission product fluorides boil at temperatures over 200°C and there are no really pertinent compounds whose boiling point is close to that of UF_6. Decontamination factors of 10^7 for both β and γ activity have been demonstrated for distilled UF_6.

A preliminary study of the technical problems associated with carrying out this fluorination and distillation in the presence of the graphite matrix material has been carried out by Atomics International (43). The following conclusions can be drawn from this work:

1) Essentially complete recovery of uranium is possible.
2) UC_2 reacts much more rapidly (violently) than does UO_2.
3) Pretreatment with Br_2 eliminates the violence associated with the UC_2 reaction.
4) The residual graphite is a potential hazard since one or more of the carbon compounds $C_nBrF_3 (n \geq 1)$, (CF), or CF_4 formed during the BrF_3 treatment decomposes spontaneously.

Additional development work is required on all the phases of this operation before it can be thought of as being a practical industrial operation.

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A significant advantage of the halide volatility process, if fuel is to be returned to the AEC, is that it is already in the chemical form required and the costly conversion of $UO_2(NO_3)_2$ through UO_2 to UF_6 is eliminated. On the other hand, if the fuel is to be reconstituted directly rather than returned to the AEC, then the advantage may be with the various nitric acid processes.

In summary then the technical feasibility of all the basic operations involved in a reprocessing plant to handle graphite-uranium fuel elements have been demonstrated on a laboratory scale. The economic significance of the several process variables remain to be studied on a pilot plant scale.

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Section 2.0

Research & Development Program

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2.0 Research & Development Program

The research and development program for any nuclear reactor type is a continual program starting with the early work which establishes basic feasibility and extending on through the operation of a number of full scale plants. The logical research and development program proceeds through a number of steps which start after a design study has shown the potential advantages of that reactor type. The initial step consists of a number of inexpensive, one-shot experiments aimed at questions related to the basic feasibility. This is followed by a number of screening tests related to major problem areas. Most often, two of these areas involve fuel elements and corrosion problems. In-pile capsule-type tests are amongst the tools used in the screening tests. The next step involves the engineering development of components which may include items ranging all the way from the complete development of new and novel devices through the performance testing of manufacturers' standard items. An out-of-pile engineering scale loop is often constructed to test components. The next step is the construction of an in-pile loop which can duplicate flow, pressure, and the temperature conditions and approach the flux level of the design study.

At this stage, a reactor experiment is usually considered. It will have a thermal rating of between 1 and 10 MW and its primary objective will be a gross demonstration of certain reactor problem areas. These may include reactor stability, fuel element performance, corrosion and component performance under irradiation.

Once reactor feasibility has been established by the reactor experiment, a reactor prototype can be built. Its thermal rating will be between 5 and 50 MW and its primary objective will be to obtain engineering information that will permit scale up to a full size nuclear power plant. Often, the reactor prototype will generate modest amounts of electricity so that an integrated power plant can be tested. There are obviously variations in this procedure, depending on time, money, and the degree of success in the various steps.

Problems associated with a reactor concept fall into two broad categories, namely (1) those problems which relate to the basic feasibility of the concept and the failure to find a solution would entail abandonment, and (2) those areas that relate to the confirmation or improvement of design in which the failure to find a solution can be readily circumvented by other approaches.

The two most significant questions for the Pebble Bed Reactor are the performance of the spherical uranium-graphite fuel elements and the operation and maintenance of the primary loop containing fission products which may leak from the fuel elements. At this time, we believe that only the question of fuel element stability belongs in the category relating to basic feasibility. If the fuel elements were to crumble or break in excessive quantities due to the combined effects of handling, gas flow, temperature and radiation then the Pebble Bed Reactor concept must be considered unfeasible. However, we do not believe that the question of circulating radioactivity in the primary loop must also be put in the first category. Actually, most other reactor concepts have this problem in varying degrees, i.e. induced activity in coolant, induced activity in corrosion products, or circulated fission products, as in the HRT and LMFR. Development work on these other promising reactor concepts should be of direct benefit to the PBR.

The research and development program for the Pebble Bed Reactor is affected by the choice of materials for the system. For example, it is felt that the selection of graphite, a good high temperature material, as the moderator and fuel element matrix and the selection of helium, an inert gas, as the coolant results in a system which inherently requires a smaller research and development program than any other combination of materials. The costly corrosion research programs of all other reactor concepts are largely avoided in this case.

The following sections outline the various efforts that would be pertinent to the R & D program for the PBR. A discussion of the current Commission-sponsored R & D programs on gas-cooled reactors and their relationship to the PBR program is given. Finally, our present recommendation of the optimum R & D program leading to the construction of a full scale Pebble Bed Reactor is given.

2.1 Fuel Elements

As discussed in section 1.0 there are three basic types of uranium-graphite fuel elements: impregnated, admixtured and lumped. Each of these types has its particular advantage for the Pebble Bed Reactor. For each type of fuel element there are a number of possible variations in materials and methods of manufacture. A systematic survey of these variables should be undertaken in order to supplement existing information. The first objective will be to produce a fuel element that will meet the performance requirements for the 125 eMW plant described in Parts I and II. Fuel elements of the various types can be readily obtained from a number of manufacturers. A continual comparison of these various types will be made with the emphasis shifting towards the most promising type. As a further part of this work, studies should be made of refabrication methods for radioactive fuel in order to take advantage of the economies afforded by only partial fuel reprocessing.

Fuel element specimens will be evaluated both with and without the presence of radiation.

2.1.1 Non-Radioactive Test Program

The pertinent physical properties of the various fuel element types should be determined as a function of temperature. These would include tensile and compressive strength and thermal conductivity. It would be desirable to duplicate the expected temperature gradients within the fuel elements by induction heating although it is known that plain graphite has the ability to plastically deform and thus relieve the stresses caused by thermal strains. Methods of either rapidly heating or cooling the specimens should be investigated in an attempt to duplicate the thermal shock that may be encountered in reactor operation.

Although uniformity of fuel dispersion is not an important requirement for the spherical uranium-graphite fuel element, radiographs should be taken to show the fuel disposition as a function of the various manufacturing techniques. Variation in loading for a number of similar elements should be noted.

Specimens should be heated in a vacuum furnace to various temperatures up to and beyond the maximum anticipated operating temperature of 2500°F. The types and quantities of any off-gas should be noted. The specimens can also be analyzed for conversion of UO_2 to UC_2 and for migration of uranium. A number of fuel elements which are compressed together can also be heated to determine if there is any tendency for the fuel element to self weld.

Relative surface stability can be determined by exposing the fuel elements to a high velocity gas stream with a filter located downstream. This test can be performed in the as-received condition, after machining dust has been removed and after the ball has been bounced or impacted a number of times. A further surface test is to determine the rate of weight loss as a function of temperature and moisture content in a helium stream.

2.1.2 Radiation Testing

There are three general types of in-pile radiation tests that can be used to test PBR fuel elements. The first is a static capsule irradiation where all information is gained from post-irradiation examination. The second type is one wherein one or more properties of the fuel element are determined while it is under irradiation. An example of this type would be the passage of sweep gas over a fuel element specimen in order to study fission product leakage. The third type of test involves an in-pile loop in which the coolant flow, temperature, and flux approximate the reactor design conditions.

The fission rate in the reference fuel element can be closely approximated by capsule tests in any of a number of presently operating research reactors. By replacing the ThO_2 with fully enriched UO_2, the fissionable material content of a specimen fuel element can be increased by a factor of 10 while at the same time maintaining the graphite-metal oxide proportions. Another factor which tends to increase the fission rate in the specimen relative to the reference design is the higher fission cross-sections due to the lower moderator temperatures in research reactors. For example, a 1-1/2" diameter specimen containing 10 w/o enriched UO_2 would generate approximately 2 KW in a 10^{13} thermal flux. This heat can be dissipated from a static capsule in a water-cooled research reactor with fuel specimen temperature below 2000°F. Screening tests of one week at this condition would produce about 4×10^{19} fissions in the specimen. This is about 4% of the fissions which occur in a fuel element exposed to the maximum flux during the reference core life. It should be noted that the most promising fuel element specimens need only be irradiated in 10^{13} flux for about six months to fully duplicate the reference design.

Many significant tests can be economically performed on the capsule irradiated screening specimens. A number of the non-radioactive tests can be repeated such as compressive strength and impact strength. In addition, weight loss and dimension change can be noted and a qualitative analysis of fission products and uranium that may leak into the capsule can be made. Since the fission rate in the capsule closely duplicates the reference design, this is the best method of duplicating internal heat generation and therefore the best method for determining the effects of thermal stress.

The types and quantities of fission products that may diffuse from the fuel elements will affect the operation and maintenance of the primary loop and fundamental knowledge of this information is desirable. This type of information can be obtained either by irradiating the specimens with subsequent analysis of the heated specimen in a hot cell or by passing sweep gas over a specimen during irradiation and performing a simultaneous analysis of the sweep gas. The former technique has been used to provide qualitative results but significant differences have been found when going to the latter technique. The latter technique is recommended for the PBR program because it will give results under the simultaneous conditions of temperature, neutron flux and fission product decay.

An additional benefit of the latter technique is that it can be used to get some information on the deposition and removal of fission products in the primary loop. The sweep gas can be brought out through materials similar to those in the PBR and in which the 1200°F to 500°F temperature gradient is maintained. After operation of the experiment, the piping can be sectioned in a hot cell and analyzed to determine the nature of any preferential deposition of fission products. The specimens can also be used to determine the efficiency of various decontaminating liquids including a simple water rinse up to and including acid etching.

The ultimate in in-pile testing would be the simultaneous irradiation of a significant number of fuel elements in an in-pile loop. The out-of-pile portion of the loop would contain a blower to recirculate helium at reference design velocities and a heat exchanger to dissipate the heat generated in the fuel elements. The objectives of such a loop, aside from furnishing irradiated fuel elements for further analysis, would be a dynamic test of fuel element performance. The circulating gas stream would be periodically sampled and analyzed for the presence of fission products, uranium and dust. A trap for removing chemically active fission products (see Part I, Section 4.3.6) would be tested in the loop. Information could be obtained concerning fission product deposition throughout the loop and its subsequent decontamination. The minimum size of the in-pile portion of the loop would be a 3" pipe containing about 10 fuel elements, which could fit into most research reactor holes. A larger section of about 12" diameter and containing about 1000 fuel elements could be rigged up in a swimming-pool-type reactor with sufficient conventional fuel elements surrounding the 12" container to make a critical assembly.

2.1.3 Fuel Reprocessing

The uncertainties associated with reprocessing have been discussed in Section 1.4. It is proposed that a systematic study be made of the reprocessing characteristics of the several alternate fuel types coupled with the alternating reprocessing schemes. It is intended that this study be aimed at uncovering and solving those problems associated with reprocessing on a commercial scale rather than the laboratory scale work that has been done heretofore.

2.2 Core Studies

There are a number of items relating to the mechanical design of the Pebble Bed Reactor core which must be confirmed or optimized before operating the reactor. These relate to gas flow, ball flow, and core components.

As discussed in Section 2.0 of Part II, there are variations of the order of ± 25% in the data reported on the friction factor, f, and the heat transfer factor, j, for flow through randomly packed beds of spherical particles. The bulk of this data has been obtained on spheres below 1/2" dia. and in a Reynold's number range below 5000. Since we are concerned with 1 1/2" dia. spheres and Reynold's numbers up to 25,000 it is important to verify f and j in this range. A variation in f is perhaps more significant than in j because core pumping power is directly proportional with f while a 25% decrease in j would only increase fuel element temperatures by about 100°F. Pressure loss data can be readily obtained by circulating room temperature air through a bed of dummy fuel elements. Heat transfer data would have to be obtained by mounting special heating elements and thermocouples within a dummy fuel element.

The non-uniform velocity distribution in the core channels (see Part II, Sect. 2.1.3) is caused by the relatively higher bed voidage adjacent to the graphite channel walls. Fortunately, this results in higher gas velocities in the region where the effect of flux peaking in the graphite walls causes the highest power density. It is difficult to extrapolate existing experimental work on velocity distribution in finite granular beds to the present core because of the large number of variables such as particle size and shape, bed diameter, and radial temperature distribution. A reactor core channel mockup can be tested by a number of methods such as velocity probes or transient temperature measurements in a preheated ball bed in order to determine the velocity distribution.

Fuel is loaded and unloaded in the Pebble Bed Reactor in a "random" method as contrasted with the precise fuel handling mechanisms of all other fixed fuel reactors which accurately locate the fuel. Therefore a core mockup should be constructed which can be repeatedly loaded with dummy fuel elements in order to study fuel handling. This mockup can be used to determine the average packing fraction and its deviation. Ball flow patterns through the core can be studied and the minimum practical angle of the bottom grates can be determined. Graphite grates can be installed and wear tested by repeated ball loadings. The ball unloading valve can be proof tested. Loading and unloading ball metering chambers can be calibrated, particularly if a full scale mockup is used. If further studies indicate that a continuously loaded pebble bed core shows promise, the core mockup can also be used to study ball flow patterns for this case.

In order to establish appropriate specifications for helium purity and the graphite moderator, a test facility should be set up to circulate hot helium over graphite. The maximum allowable helium impurities can be established. Impurities coming from graphite outgassing can be detected by sampling the helium stream.

The control rods and their drives are discussed in Section 4.2.2 of **Part I.** The control rod material is Haynes-25 alloy which contains approximately 50 w/o Cobalt. Radiation tests should be performed on this material to determine its physical properties as a function of radiation damage. A typical control rod drive mechanism should be proof tested in a mockup consisting of a control rod channel, a control rod, and mounting provisions at the top for the drive mechanism. The mockup would be instrumented to permit measurements of acceleration, rod velocity, and deceleration.

2.3 Primary Loop Equipment

2.3.1 Main Compressor

Although 20,000 cfm compressors for helium required for the 125 eMW-PBR have not yet been built, there is ample experience in the design, construction, and operation of compressors for other gases. Natural gas pipeline compressors have been built to handle 25,000 cfm at 940 psi discharge pressure. Other applications involve gas mixtures containing up to 85% hydrogen at 700 psi discharge pressure. There is sufficient knowledge and experience in this field to permit the aerodynamic design of this helium compressor and attain efficiencies of 80%. Many existing compressors use a carbon ring face seal to prevent leakage along the drive shaft. This seal is similar to the one suggested for the PBR. They have been used to seal hydrogen and a number of hydrocarbon gases but to our knowledge have not yet been used on helium. Based on present experience, it appears that back diffusion of the gas through the oil lubricated face seal is prevented but there is a question of the rate of inward lubricant leakage and the solubility of helium in the lubricant. These latter items affect the size of the gas-lubricant separator.

A typical seal should be built and tested which would test for back diffusion, lubricant leak rate, helium solubility in the lubricant, the effect of lubricant vapor on loop performance, and the efficiency of a gas-lubricant separator. The proper rate of clean sweep helium will be established.

One other question regarding the compressor is the effect of radiation on the motor windings and the seal and bearing lubricant. Shielding can be installed between the compressor and the motor to minimize radiation damage to the motor windings. A limited amount of internal steel shielding can be built into the compressor to protect the lubricant in the seal and bearings.

2.3.2 Steam Generator

There is essentially no R & D work that can be recommended for the steam generator short of building and operating a prototype unit. Such a prototype unit could be tested at one of two stages in the program. If a large non-radioactive equipment test were to be built, a prototype steam generator could be included. At this time, the capital expenditure

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for such a facility would appear unwarranted. The other stage at which a steam generator could be tested would be during installation of a Prototype Reactor. In this case, a separate fossil fuel heat source could be installed in order to test the steam generator before using nuclear heat. Both steady state and transient tests could be run in order to point up any trouble spots in the design. The prototype unit can be designed based on present engineering knowledge, and proper specifications for welding, cleanliness, and leaktightness would have been developed from prior loop programs.

Based on the 125 eMW study, it appears that a once-through steam generator is a potentially better design than a controlled circulation steam generator in that it has fewer shell penetrations, simplifies external piping, and minimizes water volume within the containment vessel. The major question on a once-through design is one of water flow distribution at the lower steam pressure of approximately 1450 psi in the 125 eMW-PBR. It is recommended that boiler manufacturers be requested to study and develop once-through steam generators similar to this application.

2.3.3 Miscellaneous Hardware

The miscellaneous hardware program for the PBR will cover equipment for handling helium containing radioactivity. Of interest in this category are the primary loop block valves, piping connectors, transfer compressor, moisture detector and dust separators if required. In general, there is no basic question of feasibility of helium system equipment for the PBR. Instead, this program would involve the manufacture of equipment to meet specifications followed by an evaluation at PBR conditions to determine their operating characteristics.

The 16" and 24" primary loop butterfly valves must have a hermetically sealed actuator and non-lubricated linkages operating in a helium atmosphere. Since they are not required to be leaktight across the seat, leak rates should be determined. Although an all welded primary system, which involves cutting and rewelding in order to replace equipment, has been recommended for the PBR, the use of metal gasketed quick disconnects should be investigated to determine their leaktightness and economic advantage. Transfer compressors for large volume helium systems should be capable of high volume flow rates. This will require developing a machine consisting of an initial centrifugal stage followed by several reciprocating stages. The problem of shaft leakage on the reciprocating compressor should be studied. In the case of the moisture detectors, the influence of radiation on their performance should be tested. If the results from flow testing on graphite fuel elements show that graphite dust will exceed tolerable concentrations, some form of dust separator will be required. This would take the form of either a filter, a cyclone separator, or an electrostatic precipitator. There is a question of basic feasibility regarding the electrostatic precipitator for this application, however it offers a unique method of both dust and fission product removal which should be investigated.

2.4 Refinement Studies

The reference design of the Pebble Bed Reactor is a 125 eMW plant operating on the Th-U 233 cycle. The entire core is batch-loaded on a 100 day cycle. There are a number of investigations that can be made to determine if this reference design can be improved.

The Pebble Bed Reactor concept is amenable to continuous refueling. In this case, no excess reactivity is required at startup and hence there will be essentially no neutrons lost to the control rods. This will result in an improved breeding ratio. Since fresh fuel must be loaded onto the top face of the core and spent fuel discharged from the bottom of the core, there will be an axial fuel concentration gradient. However, since the highest fuel concentration will be at the coolant inlet end, peak fuel element temperatures should be lower than in the case of the reference design. Sufficient work should be done to establish the fuel loading, breeding ratio, and the effect on fuel cycle costs.

As discussed in Part I, Section 1.0, there is lack of good information on the non-fission capture cross-sections for U233 (i. e. the value of α, the ratio of non-fission capture to fission capture) in the epithermal range. Values of α used for the reference design were estimated from existing data. These estimated values were higher than other currently accepted values. This spread was of sufficient magnitude to cause the reference design to be a true "breeder" reactor when the currently accepted α values were used and to make the reference design be merely a "converter" when the new estimated values were used. Since all other power reactor types employing the U233-Th cycle can be affected by this uncertainty, it is recommended that the Commission undertake basic research to clear up this matter.

The performance of the PBR using partially enriched uranium should also be established. Studies would include the degree of enrichment required for the present reference design using a depleted uranium blanket, core size for a low enrichment fueled reactor, and the application of the seed-blanket concept to the PBR. The effect on cost of electrical power would be determined relative to the present reference design.

2.5 Relation to Other Gas-Cooled Reactor R & D Programs

There are at least two programs that are currently being supported by Commission funds that could furnish information relevant to the Pebble Bed Reactor program. One is the Gas-Cooled Power Reactor Project at the Oak Ridge National Laboratory and the other is the Process Heat Reactor Program being performed under a cooperative program by the Commission and the Branch of Coal Gasification of the U. S. Bureau of Mines.

The Bureau of Mines Program is concerned with the development of a reactor using spherical uranium-graphite fuel elements. Helium is heated from a reactor inlet temperature of 1000°F to a reactor outlet temperature of 2500°F. This program is similar to the PBR in many respects. The major difference is the higher operating temperatures of the Process Heat Reactor. Their present work involves the development of the primary loop equipment and non-nuclear testing of components, including fuel elements. An atmospheric pressure helium loop has been in operation for nearly a year. Induction heating has been used to heat a variety of graphite and clad-graphite spheres. A 250 psi loop is currently under construction. Pertinent areas of interest to the PBR include:

1. Erosion and contaminant corrosion in a helium graphite system at 2500°F.
2. Heat transfer and friction factors for helium flow through ball beds.
3. Evaluation of clad graphite spheres.
4. Development of dust removal equipment.
5. Performance of equipment handling high pressure, high temperature helium.

The ORNL Gas-Cooled Reactor Program is based on the use of slightly enriched UO_2 pellets encased in stainless steel as the fuel element. The reactor is graphite moderated and uses helium as the coolant. Reactor inlet and outlet temperatures are 460°F and 1000°F. Experimental work is currently under way at ORNL and Title II design on a 40,000 KW Prototype is being performed by Kaiser Engineers -ACF.

The areas of common interest with the PBR program involve the effect of contaminants in the helium graphite system and the testing of primary loop equipment. Work is under way on the rates of adsorption and evolution of gases by graphite and on the reactions of graphite with contaminants. Three facilities are being used in this work. These include a static materials test, a thermal convection loop to test the effects of a temperature gradient, and a forced convection loop to determine if there are any erosion effects. This work will be followed by an in-pile loop to test the effect of radiation. ORNL has also proposed to study the radiation damage to pressure vessel materials. Experience gained in the manufacture and testing of primary loop components such as the helium compressor and steam generator would also be pertinent to the PBR Program.

2.6 Recommended Program

The objective of the research and development program recommended for the Pebble Bed Reactor is to bring it to a state of development which will permit the design, construction and operation of the reactor system for central station use. Since any R & D program will continually shift in emphasis as it proceeds, our recommendations here are confined to a Phase I, covering fuel element technology and Title I work on an experimental reactor.

One of the principal questions raised in past proposals of this reactor concept has been that of fuel element stability under irradiation and a "quick and dirty" reactor experiment has been proposed as a means of demonstrating this fuel element characteristic. We are now of the opinion that the experimental reactor should be used to answer problems relating to the operation and maintenance of a hot primary loop as well as fuel and fuel handling. Its construction should be preceeded by an orderly program of fuel element development in order to reduce uncertainties regarding the fuel element. Subsequent development work on such items as fuel handling and component testing would be done within the established framework of the experimental reactor. It is felt that this program can best provide the information required to permit the construction of a large power reactor.

Continuing engineering analysis of the power plant system can and should proceed concurrently with the initial R & D program, since such analysis will place individual parts of the R & D program in proper perspective with relation to the overall objective. Also, other Commission programs should be continually monitored to avoid duplication of effort.

Following is an outline of the recommended research and development program for the first phase. A time schedule is not established here as it will depend on the rate of progress of work under the present contract, under related Commission sponsored programs and the degree of acceptance of this reactor concept by the Commission and the Power Industry.

A. Fuel Element Development

The objective of the fuel element development program is to select the most promising fuel element from among the many possible types and subject it to sufficient tests that will minimize the probability of its

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failure in an experimental reactor.

1. Screening tests

A series of pre-irradiation tests would first be performed on a large number of specimens in which the method of adding fuel to graphite, fuel loading, fuel particle size, graphite type, etc. are varied. Tests would include tensile, compressive, and impact strength, thermal conductivity, abrasion resistance, moisture and oxidation resistance, and fuel particle stability.

Following these tests, a series of short time, high temperature irradiation exposures would be made followed by longer exposures on the more promising types. Static exposures in a relatively simple capsule would be employed. Many of the pre-irradiation tests would be repeated after exposure. Additional tests would include dimensional change, weight loss, and analysis of capsule gas space and walls of capsule for type and quantity of fission products and fissile material.

2. Fission product technology

A relatively simple in-pile test would be performed using sweep helium over a fuel element. This loop would be used to study the rate of fission product leakage, the nature of fission product deposition, and methods of decontamination. This system could also be used to study continuous fission product removal equipment for both the chemically active and the noble gas fission products.

3. In-pile loop tests

A recirculating in-pile loop will be used to duplicate temperature, velocity, and heat flux of a full scale reactor. Only the most promising fuel elements would be tested under these conditions. This loop would represent the final test of fuel elements prior to their use in an experimental reactor.

4. Manufacturing and reprocessing

Since the methods and costs associated with fuel element manufacturing and reprocessing are involved in the selection of the optimum fuel element, work in these areas would proceed concurrently with fuel element testing. As results of fuel element testing are obtained, emphasis on manufacturing methods will shift towards the more promising types. An evaluation of possible methods of refabricating radioactive fuel elements will also be made. As presently seen, reprocessing methods will be essentially independent of the ultimate fuel element except for the possible effect of coatings. Reprocessing methods which would be investigated include incineration, crushing and leaching, and BrF_3 volatility.

B. Experimental Reactor

In order to establish the framework for component development and testing, the Title I design of an experimental reactor should be scheduled to proceed in parallel with the fuel element development program. At this time it appears that Title I work can commence at the completion of the initial fuel element screening program. Based on the results of this work, which would establish the size and scope of the experimental reactor, component development and testing would be undertaken. This work would include gas flow thru ball beds, ball handling techniques, testing of reactor components such as the unloading valve and control rod drive and primary loop components such as blowers and valves.

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