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THE PEBBLE BED REACTOR PROGRAM

Current Fuel Element Developments and their Effects on the Pebble Bed Reactor Development Program

May 10, 1960

Work Performed Under AEC Contract AT(30-1)-2207

& AT(30-1)-2378

SANDERSON & PORTER

NEW YORK, N.Y.

DISCLAIMER

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Abstract

Recent fuel element irradiations have demonstrated the structural integrity of spherical uranium-graphite fuel elements at burnups in excess of the design requirements of a 125 eMW PBR power plant. Other irradiations indicate the successful development of a coated fuel particle which permits fabrication of fuel elements meeting the PBR design objectives of a fission product release rate (R/B) $\leqq 10^{-6}$.

β plus γ system activity in a 125 eMW PBR is 490 curies, assuming complete release of 10^{-6} of all fission products volatile at or below 2500°F. The low R/B being obtained from PBR fuel elements indicates that decay during diffusion of the short-lived volatile precursors of non-volatile daughter products will result in further reduction of this system activity, and an increase in the average half-life of the fission products remaining in the system will increase the efficiency of a by-pass cleanup system.

The method of fabricating coated particles by the hydrolysis of metallic chlorides to produce oxides or pyrolysis of hydrocarbons to produce carbon on a suitable substrate, is described and pre-irradiation test results are given.

An accelerated coated fuel particle program is discussed as well as development work on the Pebble Bed Reactor concept as a whole.

1.0 Matrix Irradiations

SP-4 is a static capsule containing six different uranium-graphite fuel elements which were irradiated in the Battelle Research Reactor from August 26, 1959 to March 7, 1960. Total exposure was 3350 hours and the average burnup of the six elements was 6700 KW Hrs per ball, equivalent to 6.7% atom burnup or 53,600 MWD/ton of contained uranium. Description data on these elements are given in Table I. Pre- and post-irradiation characteristics are given in Table II.

While the coating of the two coated elements cracked, the irradiation demonstrated the suitability of the uranium-graphite matrix as a PBR fuel element within current design requirements.

TABLE I

Specimens Irradiated in SP-4

Number	FI-1	FA-2	FA-1	FA-10	FA-8	FA-20
Mfg.	Syl.	BMI	NC	GLC	Carbo	NC
FUEL						
Loaded as	UNH	UO_2	UO_2	UO_2	UC_2	UO_2
Final Form	UO_2	UO_2	UO_2	UO_2	UC_2	UC_2
Particle Size, μ	1	67	100/150	350/420	100/200	100/150
MATRIX						
Reimpreg.	no	no	no	yes	no	no
Net Density	1.65	1.63	1.62	1.80	1.63	1.65
Bake Temp., °F	———	2000	2560	2000	3600	4800
COATING						
Material	———	———	———	———	SiC-Si	Pyro-C
Thickness	———	———	———	———	.030"	.002"

TABLE II

Pre- and Post-Irradiation Characteristics of SP-4 Specimens

Number	FI-1	FA-2	FA-1	FA-10	FA-8	FA-20
Weight Change,%	-0.40	-0.55	-0.11	-0.06	+0.14	+0.19
Dimensional Change,%						
Polar	-0.76	-0.03	-0.47	-0.69	-0.16 to	-0.93 to
Equatorial	-0.67	-0.08	-0.52	-0.81	+0.65	-0.17
Impact Failure, Ft Lbs						
Pre	12.5	7.5	>11.6	10.4	———	———
Post	>11.6	3.0	9.4	>11.6	———	———

2.0 Fission Product Retention

A spherical uranium-graphite fuel element containing Al_2O_3 coated fuel particles is being irradiated in the Battelle Research Reactor in a flux of approximately 2×10^{13} and at a ball surface temperature of 1360°F. This element was fabricated by National Carbon and fueled with Al_2O_3 coated fuel particles made by BMI. Descriptive data on the element is given in Table III.

Capsule SP-5 completed its second two week reactor cycle as of May 2nd and is presently half way through the third cycle. Five samples of sweep gas activity have been taken, during which the R/B of five selected long-lived isotopes were determined. Results are given in Table IV. R/B is averaging 10^{-7} at a ball surface temperature of 1360°F and a power output of 1.5 KW/ball. Accuracy of these data is judged to be within half an order of magnitude.

These results indicate the potential effectiveness of an Al_2O_3 fuel particle coating as a fission product barrier in a uranium-graphite fuel element. If comparable results are obtained after irradiation of 5000 KW Hrs. per ball, the major objective of the Pebble Bed Reactor Fuel Element Development Program will have been accomplished. If for any reason these particles fail, as indicated by a rapid increase in R/B during irradiation, further irradiations will be made in Capsule SP-6 with particles presently being manufactured, designed to improve performance in general.

TABLE III

Characteristics of Coated Particle Element
Irradiated in Capsule SP-5

Number	FA-22
Mfg.	NC/BMI
Fuel	
Loaded as	UO_2
Final Form	UO_2
Particle Size,	100/150
Matrix	
Reimpreg.	no
Net Density	1.57
Shell thickness, in	———
Bake temp., °F	2350
Coating	
Location	Particle
Material	Al_2O_3
Thickness, in.	.002

TABLE IV

Fission Product Release Rates R/B of SP-5 Specimens

Specimen	FA-22				
Days	5	9	19	22	23 (1)
Surface Temp., °F	1160	1160	1360	1360	1360
Power, KW/ball	1.3	1.3	1.5	1.5	1.5
Kr 85m - $t^{1/2}$ = 4.36 hrs.	1.0×10^{-8}	3.6×10^{-8}	2.9×10^{-7}	4.6×10^{-6}	1.6×10^{-7}
Kr 87 - $t^{1/2}$ = 78 min.	$< 1.0 \times 10^{-9}$	8.2×10^{-9}	8.2×10^{-8}	(2)	(2)
Kr 88 - $t^{1/2}$ = 2.77 hrs.	1.0×10^{-9}	1.2×10^{-8}	1.3×10^{-7}	(2)	7.5×10^{-9}
Xe 133 - $t^{1/2}$ = 5.27 days	8.3×10^{-8}	4.7×10^{-8}	8.5×10^{-7}	2.6×10^{-6}	5.7×10^{-8}
Xe 135 - $t^{1/2}$ = 9.13 hrs.	9.0×10^{-9}	1.8×10^{-8}	2.2×10^{-7}	9.3×10^{-7}	3.7×10^{-9}

NOTE: (1) Preliminary data

(2) Activity present but not measured.

3.0 PBR System Activity

It has previously been determined [1] that the total γ activity in the primary loop assuming complete release of all fission products volatile at or below 2500°F, is 2.3×10^8 curies or 1.1 mC/cc (NTP) of design system volume. To this must be added 2.6×10^8 curies or 1.3 mC/cc of β activity due to fission products. (This does not include the β activity due to tritium). At that time it was suggested that an R/B of 10^{-6} might be realized. This reduction in leakage rate was applied "across the board" and resulted in a γ activity in the primary loop of the 125 eMW PBR design of 230 curies and a β activity of 260 curies (exclusive of tritium which is not influenced by R/B).

The fission product release data presented in Table IV indicate that, for the long-lived gaseous fission products: Kr 85m, Kr 87, Kr 88, Xe 133 and Xe 135, whose γ activity represent 20% of the total, an R/B of the order of 10^{-6} is possible. A gradual increase in leakage rate, at both temperatures at which readings have been taken, is to be noted. This raises the question as to whether the coated particles are experiencing progressive failure due to radiation damage and/or gas pressure buildup or whether the increase is due to the diffusion mechanism. Increase in R/B due to the first cause can only be speculated on as there is no data available on irradiation damage to α Al_2O_3. Gas pressure buildup leading to rupture of the coating can probably be ruled out since rupture of the coating on even

(1) NYO 2373 Section 5 pp. 5-12.

a single particle would result in a total R/B at least one order of magnitude higher than has been observed. Increase in R/B due to the diffusion mechanism is more readily explained, since diffusion through a barrier is dependent upon the concentration gradient of the diffusing components, which in this case is time dependent. A continuous increase in R/B can be expected until such a time as the leakage and decay rate are in equilibrium with the production rate.

This latter point is extremely important for from it follows the fact that the fission products which do ultimately leak are relatively old. Short half life materials will have decayed within the barrier and consequently need not be reckoned with in considering PBR system activity. Or saying this another way, the R/B for any one chemical species is expected to vary directly with half life. The relationship between half life (decay constant) and R/B is shown in Figure 1 for a range of diffusion constants from 10^{-13} to 10^{-7}. Also shown on this figure are the decay constants for fission products being measured (Table IV) and the mass numbers of short half life gaseous precursors of long lived solids which are important from the point of view of shielding and maintenance. Thus the mark indicating Ba-La 140 is plotted at $\lambda = 4 \times 10^{-2}$, corresponding to the half life of Xe 140, and indicates that if an R/B of 10^{-6} is measured for Xe 133 for example, the effective R/B for the chain headed by Xe 140 will be as low as 10^{-41}.

If it is assumed that the chains whose gaseous precursors have half lives less than 60 sec are eliminated completely, the total γ activity will have been reduced to 74% of its former value. Applying an R/B of 10^{-6} to the remainder would result in a gross γ system activity of .9 mC/cc.

The present sweep capsule SP-5 is fitted with a short lived activity trap which will be sampled at a propitious time to determine what short lived fission products, if any, are being released from the fuel elements being irradiated. We are awaiting the results of this series of tests for confirmation of our assumptions concerning the holdup of short half-lived precursors and the consequent reduction in overall system activity.

Another significant point is that as the average half life of the fission products released increases, the efficiency of a bypass fission product removal system increases. This is indicated in Figure 2 where it can be seen for example, that an increase in the average half life from 2 to 19 hours, increases the decontamination factor one order of magnitude, for the same bypass flow.

Tritium activity, which activity is in no way related to fission product release and is a part of every helium cooled reactor being contemplated, is being added to the PBR system at a rate of about 11.5 curies per day. Equilibrium tritium activity will not be reached in any reasonable time, hence the activity can be considered as growing more or less constantly. Since this is a soft beta emitter, there is no radiation

hazard external to the primary loop. There is however the requirement that tritium in air be no more than 7×10^{-5} μ C/cc. Hence if the PBR was operating for say 3 years and its coolant had to be released to the atmosphere, it would have to be diluted with 10^5 times its own volume of air to reduce the tritium activity to tolerance.

Such a dilution factor would be adequate in reducing all other activity to tolerance even if the impractical assumption is made that none of this remaining γ activity plated out on the inside walls of the primary loop. Actually it is not intended that this primary loop helium be released to atmosphere. At the time of a shutdown for maintenance, it is intended to simply transfer the helium to storage flasks. The residual activity within the system will depend on the amount and kind of plate-out which has occurred. An in-pile loop is currently being constructed for operation at BNL to evaluate this latter point.

4.0 Coated Fuel Particle Development

Four types of coated fuel particles are being developed, or planned for development, under the present Fuel Cycle Development Program. They are: vapor-deposited oxides, Al_2O_3 and BeO, a vapor-deposited carbide, SiC-Si, and pyrolytically-deposited carbon, PyC. Characteristics of these types of materials, together with those of fissile and fertile materials which would be coated, are given in Table V.

Several benefits are derived from the fabrication of PBR elements with coated particles. With regard to fission product retention, the coating is at the source and in a properly made element the danger of cracking a coating is eliminated. If a poor matrix gets through inspection and breaks during irradiation in the reactor, fission products released, if any, would be several orders of magnitude less than what would be released from an element with a surface coating. The number of coated fuel particles in a reference PBR element is $980,000/D^3$ where D is in $\mu/100$. Thus, the number of coatings to retain fission products in an element fueled with 200μ particles is 125,000, in contrast to a single coating in a coated element. The total γ activity associated with a single broken particle is but 9 milli curies where the total γ activity associated with a broken element is 1145 curies, assuming complete release of all fission products born.

Preliminary cost estimates indicate that the production of particle coatings will be less expensive than the production of element coatings.

TABLE V

General Characteristics of Fuels & Cladding Materials

Material	Fuel Particles			Coatings			
	UO_2	UC_2	ThO_2	Al_2O_3	BeO	PyC	Si-SiC
Density, theoretical, gm/cc	10.96	11.68	10.50	3.97	3.03	2.26	2.7
Melting Point, °C	2750	2475	2050	2050	2530	3700	1425 (1)
Reaction Temp., with carbon, °C	1500	——	1950	1800	2300	——	1200 (2)
Thermal conductivity, cal/(sec)(cm)°C	0.014	0.080	0.007	0.019	0.046	0.012	0.2
Expansion Coeff., 10^{-6}/°C at 1000°C	10	12.5	9	8.5	10	a-2.05 b-25.4	4
Rupture Modulus, psi	18,000	20,000	7,000	100,000	23,000	17,500	20,000
Σ_a, cm^{-1}	——	——	——	.0105	.00065	.00037	.0074

(1) Si melts
(2) Upper Service Temperature

The cost of fabrication of elements with coated particles will also be less expensive than the cost with uncoated particles due to the reduction of radiation hazards.

4.1 Al_2O_3

Initial work on Al_2O_3 coatings was done by sintering a reactive grade of Al_2O_3, reported in BMI-1321 and NYO-2706 Phase I Progress Report. Results were not satisfactory and as a consequence deposition of Al_2O_3 by hydrolysis of $AlCl_3$ in a fludized bed was investigated, which resulted in a satisfactory material. In this process $AlCl_3$ is decomposed in the presence of water vapor so that Al_2O_3 molecules are deposited on the surface of the seed material, which is a UO_2 particle. Thus a dense impermeable α-Al_2O_3 coating is formed, molecule by molecule. The most important variable is temperature of deposition, although gas composition, flow rate, and bed height can also be significant. A list of the five batches of Al_2O_3 coated particles made to date is given in Table VI.

TABLE VI

Al_2O_3 Coated Particles and Fueled Spheres Made to Date

Batch No.	UO_2 Size	Enrich.	Coat Thick.	Deposit. Temp.	gms of UO_2	No. of Spheres
1	105/149μ	Nat.	20μ	1830°F	50 gm.	3
2	105/149	Nat.	38	1830	50	2
3	105/149	Nat.	50	2010	75	0
4	105/149	Enr.	55	1830	50	3
5	250/420	Dep.	40	2100	75	0

The 20 micron thick (batch No. 1) and 38 micron thick (batch No. 2) Al_2O_3 coatings on UO_2 are shown in Figures 3 and 4 respectively. Batch No. 2 was made using a portion of batch No. 1 as the starting material. As can be noted, all UO_2 particles are quite uniformly coated with dense continuous Al_2O_3. The coatings in batch No. 1 were applied in three successive separate steps. As can be noted in both Figure 3 and 4, good bonding is achieved between successive layers. The knob formations in Figure 4 are not objectionable. The cause of these knobs is not presently known, however it is interesting to note that similar formations have been observed in molecular carbon deposits (i.e. pyrolytic graphite).

Figure 5 and 6 are 100x and 500x magnifications from batch No. 4, the material presently being irradiated in Capsule SP-5. This coating was applied in 5 successive steps. A darker band is seen at the third layer. This darker region was undoubtedly caused by a shift in one or more of the process conditions while depositing the third layer. Again, all layers appear well bonded to the previous layers. More evidence of dark layers was found in batch No. 3.

The standard screening test for coating integrity is to heat the particles in air at 1200°F so that any crack in the coatings will permit the UO_2 to oxidize to U_3O_8. Thus, weight gain is a measure of the lack of coating integrity. No weight gains have been found in any of the batches to date.

Actually some slight initial weight losses have been noted, believed to be due to devolatilization of adsorbed gases.

One problem in fabrication was uranium contamination of the coating from UO_2 dust abraded from the fuel particles in the initial stages of coating. This was overcome by stopping the reaction after several microns of Al_2O_3 had been formed. cleaning the reactor, and restarting. Using this technique uranium contamination of 10^{-7} to 10^{-5} of the uranium in the particles has been achieved, as measured by a 2π gas flow alpha counter.

Due to the slightly higher thermal expansion coefficient of UO_2, compared with Al_2O_3, the possibility of coating rupture exists when the particles are heated above the deposition temperature. One thermal cycle test was run on 40-100/150μ particles (batch No. 2) in air between 2500° and 100°F. An increase in uranium contamination and microscopic examination gave definite indication of coating failure. However, when the same test was repeated in a non-oxidizing atmosphere, no failure was noted. This indicates that oxidation of the UO_2 by diffusion through Al_2O_3 may well have been the cause of the previous failure. Next, 40-250/420μ (batch No 5) particles (i.e. lower ratio of coating thickness to particle diameter) were cycled in a non-oxidizing atmosphere to 2500°F and coating failure was again detected. Thus it appears that there will be a minimum allowable ratio of coating thickness to particle diameter.

Reactions between Al_2O_3 and graphite matrix materials are being studied. Earlier results with a sintered alumina in graphite showed severe reaction at 3000°F and no reaction at 2500°F. No reactions have been found at 2500°F after 500 hrs. using vapor deposited alumina and a graphite made from graphite flour and a coal tar pitch binder. However, some reaction was noted at 2500°F when using graphites made from coke fillers and/or resin binders.

To date, a total of eight spheres fueled with Al_2O_3 coated particles have been made. Efforts to remove the molding band on one of the spheres by grinding resulted in an increased uranium contamination in that region due to abrasion of the particle coatings. Radiography of slices of the fuel element has shown a uniform distribution of the particles throughout the graphite matrix. Numerous particles could be observed just below the graphite surface.

Post-irradiation heating tests have been run both on particles and on a fueled graphite sphere. Results are given in Table VII. As can be noted, mixing of the fuel particles with the graphite materials, molding, and baking has apparently caused no damage to the particle coatings. Furthermore, a thermal cycle test between 1900°F and 600°F also caused no apparent increase in fission product release.

Furnace capsule SPF-3 has been operated at two temperatures with a graphite sphere fueled with 40 micron Al_2O_3 coated UO_2 particles.

TABLE VII

Post-Irradiation Heating Data on Al_2O_3 Coated UO_2

Al_2O_3 Coating Thickness: 40μ - UO_2 Diameter: 100 to 150μ

	Particles Only	After Admixture into 1-1/2 in Graphite Sphere			
Temp., °F	1500	1500	1900	1900[(a)]	1900[(a)]
Time, min.	144	270	60	60	135
$\frac{\text{Xe 133 Released}}{\text{Xe 133 Present}} \times 10^6$	1.3	7.5	<9.4	<9.4	<9.4

(a) Cooled to 600°F prior to these runs.

In this experiment, fission product release is measured while the specimen is under low level irradiation. Temperature is adjusted by an electrical heater. Results are given in Table VIII. These Results are somewhat higher than those obtained from Sweep Capsule SP-5. One possible explanation is the difference in coating thickness. It is planned to continue operation of SPF-3 at temperature up to 1900°F.

TABLE VIII

Fission Product Release Data on Graphite Sphere Fueled with Al_2O_3 Coated UO_2

Al_2O_3 Coating Thickness: 40μ - UO_2 Diameter: 100 to 150μ

Data from Capsule SPF-3

Sample No.	Flux	Temp. °F	R/B (release rate/production rate) Xe 133	Xe 135	Kr 85m	Kr 87	Kr 88
1	2×10^{12}	150	(No detectable release)				
2	"	1000	1.2×10^{-6}	1.7×10^{-7}	5.6×10^{-7}	4.2×10^{-7}	2.6×10^{-7}
3	"	1500	5.8×10^{-6}	1.7×10^{-6}	2.0×10^{-6}	1.0×10^{-6}	3.2×10^{-7}

A number of additional batches of Al_2O_3 coated particles are planned for immediate fabrication. Another batch of 105/149 micron fully enriched UO_2 will be coated, primarily for use in the In-Pile Loop program. A 200 gm. "production run" will be made, using 105/149 micron co-precipitated ThO_2-UO_2 (11:1) high-fired shot. In addition to investigating the use of thoria, this batch will be used for graphite sphere fabrication studies. A series of runs will also be made using a number of UO_2 sizes (from 105 to 420 microns) with various Al_2O_3 thicknesses in order to investigate thermal cycling rupture as a function of coating thickness-to-UO_2 diameter ratio.

4.2 BeO

While excellent results are being obtained with vapor deposited Al_2O_3 as a fission product barrier, it is the least attractive of the three coatings being studied, based on considerations other than its fission product retention ability. The most desirable fuel particle coating for the PBR concept is BeO. The thermal expansion coefficient of BeO more nearly matches that of UO_2, eliminating differential stress as a source of coating failure. The thermal conductivity of BeO is slightly higher than that of Al_2O_3, resulting in lower fuel particle temperatures. The reaction temperature of BeO with carbon is about 500°C higher than that of Al_2O_3, which removes any concern over temperature limitations in the PBR. BeO does not displace moderator, which is an important point as the volume of a 50μ coating on a 200μ fuel particle can displace about 5% of the moderator.

In addition the γ-n and n-2n reaction results in the release of additional neutrons which enhance the breeding potential of the PBR.

To our knowledge the technical feasibility of vapor deposition of beryllium chloride by hydrolysis on a fuel particle has not been established. Facilities are available at BMI in which this work can be done and our present schedule is to coat a first batch of particles for evaluation in mid-June.

4.3 PyC

The second most desirable fuel particle coating is pyrolytically deposited carbon. Such a coating will not displace moderator and when deposited on UC_2 would provide an element that is essentially not temperature limited. Pyrolytic carbon (PyC) is formed by the pyrolysis of a gaseous hydrocarbon on a suitable substrate at temperature ranging from 1000 to 2500°C. At the lower temperature range the material is an extremely fine-grain, dense carbon. When deposited at temperatures at and above 1800°C, the microstructure is typical for vapor-deposited material and is fibrous in nature. It is impervious to gases and liquids, is much stronger than commercial graphite and is less reactive towards oxygen. It exhibits a greater degree of anisotropy than single crystal natural graphite. Figure 7 illustrates a pyrolytically deposited carbon coating at 250x (Raytheon) which us typical of material deposited at high temperature.

A modest amount of development has been and is being done to deposit pyrolytic carbon on fuel particles as a reaction barrier with fuel element cladding materials. Our development, which was initiated in December 1959, is the only one to our knowledge designed to retain fission products. Two types of PyC coated particles are possible; low temperature deposition on UO_2 and high temperature deposition on UC_2. The dividing line is of the order of 1500°C meaning that UO_2 particles would be coated with fine-grain carbon and UC_2 particles with the fibrous fine-grain graphite. Our present development is on the lower temperature material, because of temperature limitations of process equipment. Two batches of UC_2 particles, shown in Figure 8, have been coated by the pyrolysis of methane and acetylene at temperatures ranging from 1000 to 1100°C. Material from the better of these two batches, which was a 40μ coat on 250 to 420μ UC_2 was leached, α-counted and thermal cycled. Initial leaching with nitric acid showed essentially no uranium external to the particle. An α count showed an activity of <0.7 cpm/gram of coated particles. The material was then thermally cycle three times between 2000°C and ambient. A subsequent α count showed 26,000 cpm/gram and a subsequent nitric acid leach showed a strong qualitative indication of uranium. Photomicrographs were made, shown in Figure 9, which disclosed the ruptured coatings which were anticipated on the basis of the α count and nitric acid leach. This was a most severe test and the coating potential should not be discounted because of

the adverse results. Further tests in progress on this material are: neutron activation where the release of fission products will be noted at 1500, 2000 and 2400°F heat treatment, and determination of density which is judged to be about 2.0 from inspection of photomicrographs. Further work in this area will be PyC deposition on 250 to 420μ spherical UC_2 shot at both 1000 and 1400°C and a study of high temperature deposition, i.e. in the range from 1400 to 2000°C.

4.4 SiC-Si

Siliconized silicon carbide containing an excess of free-phase silicon has proven to be one of the most effective materials examined to date for the retention of fission products. However, its application to date has been as a coating on a 1-1/2 inch sphere and test results have been unsatisfactory from a structural standpoint. This can be attributed to a number of things, all of which are yet unresolved. Capsule SP-5 contained, in addition to an Al_2O_3 coated fuel particle element, a 3M SiC-Si coated element. This element failed on going up to power at the start of the third cycle for reasons yet unknown. However, after seventeen days irradiation the R/B averaged 10^{-9} for Kr 85m, Kr 87, Kr 88, Xe 133 and Xe 135.

While no further coated element development is contemplated, a development of SiC-Si coated particles is under way on the part of one manufacturer (3M). Exploratory work has been done which has consisted of experimenting with a barrier between the fuel and coating to prevent uranium

[redacted]

contamination. In view of the failure of the 3M element being tested and the encouraging results of coated particles, we are advised that 3M will pursue SiC-Si coated particles more intensively. This is a private development.

5.0 Fuel Cycle Development Program (AT-(30-1)-2378)

Sanderson & Porter's effort in the Fuel Cycle Development Program is directed towards developing a fuel element which is satisfactory for use in the Pebble Bed Reactor. However, the development of coated fuel particles capable of retaining a high percentage of the fission products generated, makes these fuels applicable to other uranium-graphite reactors having requirements that differ from the PBR. This may indicate a more extensive process development and characterization than scheduled in our present program.

Based on the 125 eMW PBR requirements the design burnup of all fissionable atoms is about 5.5%, as controlled by reactivity. This and other PBR designs, as well as uranium-graphite reactors of other types, may raise problems of coating failure due to fission product gas pressure. Even if specimen FA-22, being irradiated in Capsule SP-5, generates 5000 KW Hrs of heat without failure, it is deemed to be desirable to irradiate particles to failure in a capsule configuration which would permit identifying specific particles for pre- and post-irradiation examination. Such particles would include, but not be limited to, ones similar to those being irradiated; particles made with low density fuel, and particles with a porous substrate to provide a residence space for fission product gases. Such irradiations would be performed in a simple uninstrumented capsule

which would be one of a group in a single irradiation. The number of particles to be irradiated would be selected so that post-irradiation examination need not be performed in a hot cell.

Immediate consideration should be given to production requirements. The laboratory reactors presently being used for coated particle development (Al_2O_3 & BeO) are limited to about 100 grams per day output. A pilot production plant having a capacity of kilograms per day should be built to explore the problems of scale-up and reproduction of labroatory quality in production quantities. This facility should be built in the immediate future and be capable of use with either Al_2O_3 or BeO, with the latter dictating the design because of hazards.

Present PyC coated particle development is restricted to the study of carbon deposition at or below 1400°C because of temperature limitations of the equipment. PyC coatings deposited between 1400°C and 2500°C should be explored to determine the characteristics of materials obtained at these temperatures.

Several problems remain in production development of completed fuel elements themselves. One is the modification or removal of the wide belt existing on balls molded in the conventional manner, in a hydraulic press. This is a manufacturing problem and calls for die development. Another more important point is to provide a handling barrier on the fueled

ball to prevent the dislodging of surface particles during handling. Such a barrier can take the form of an unfueled graphite shell, a PyC coating, or a soft carbon coat on the particles themselves which would serve to locate the center of the particle well below the surface of the ball, yet become an integral part of the matrix upon molding. A third problem is to determine the necessity of a surface coating to protect the graphite against a reaction with oxygen (exothermic) or a reaction with steam (endothermic). The reaction rate of selected PBR element matrix materials should be determined and if the rate is unacceptable, consideration should be given to a surface coating, such as siliconized silicon carbide.

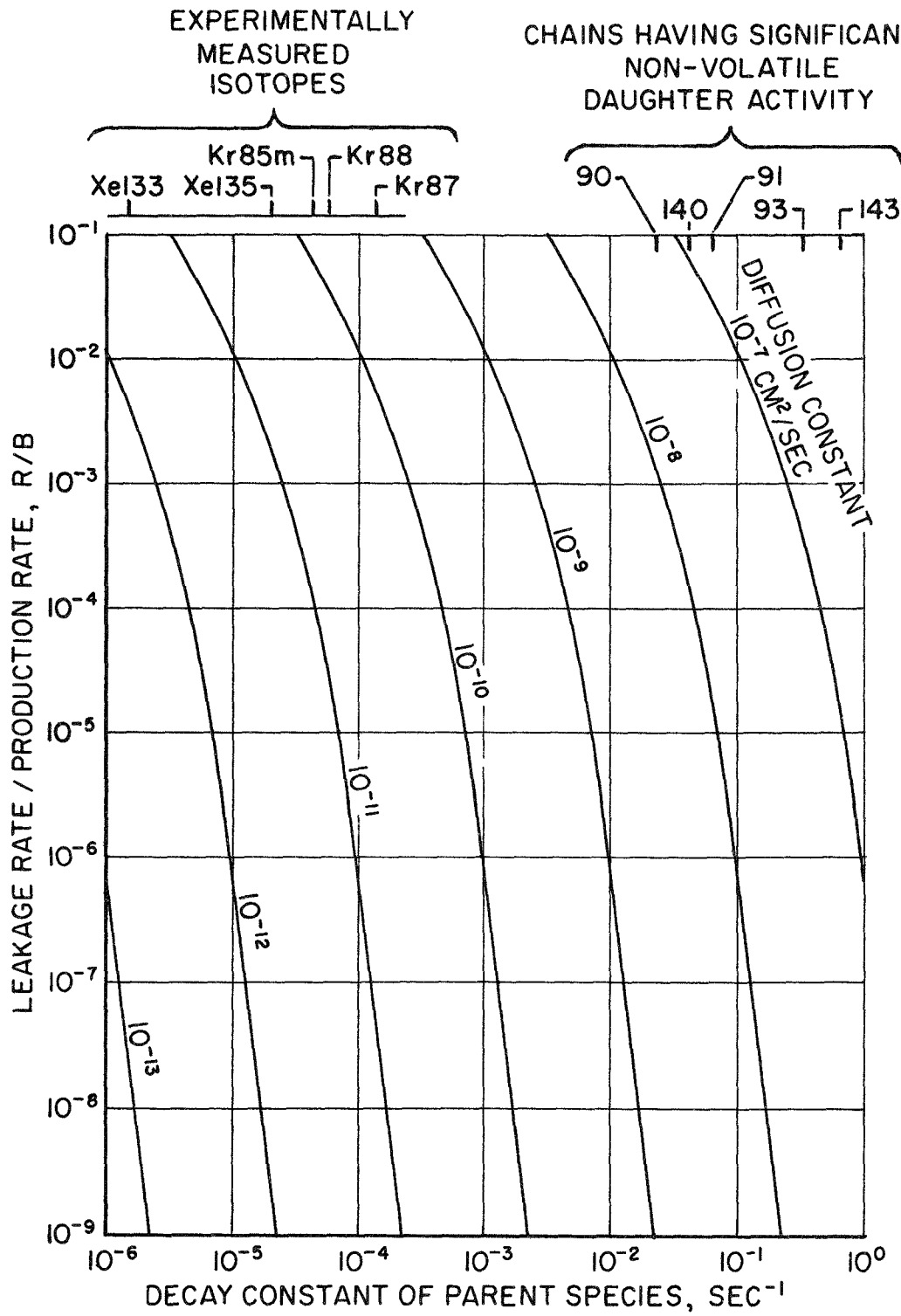
The continuously loaded PBR is unique among reactor types in that fuel discharged for reprocessing has been irradiated to the maximum allowable burnup. The full economic advantage of this type reactor cannot be realized unless continuous reprocessing of the daily discharge of fuel is practiced. One of the problems in reprocessing uranium-graphite fuel elements has been that of handling large quantities of activated graphite. This problem is not so severe with coated particle elements. A study should be made of the economics of on-site reprocessing, where the load is some 70 Kg per day of fuel elements. This would include a preliminary design; capital and operating cost estimate of a head-end and fission product separation plant taking into consideration decontamination factors; and, final determination if one or both are required at the site to effect maximum economics in reprocessing spent fuel from the PBR.

6.0 Pebble Bed Reactor Program - (AT(30-1)-2207)

Report S&P 1965-3 recommends building a reactor experiment to demonstrate the feasibility of the Pebble Bed Reactor concept by a) bulk irradiation of fuel elements b) maintenance of activity at specified levels by cleanup of a by-pass stream c) use of feasible maintenance procedures, and while not related to the basic feasibility of the concept d) demonstrate reloading procedures during reactor operation. These recommendations were made on the assumption that the fuel elements would have a surface coating capable of retaining all but 10^{-6} of the fission products born. The use of coated particle elements does not obviate the necessity of irradiating a bulk quantity of fuel elements to demonstrate feasibility. However, the need to demonstrate maintenance of system activity at specified levels diminishes with lower release rates and problems of system maintenance are lessened. It is still desirable to demonstrate reloading procedures, under load. An experimental reactor program has been repeatedly put back, awaiting favorable results from the Fuel Cycle Development Program. In view of the present results it is felt that its design and construction can now be started with confidence.

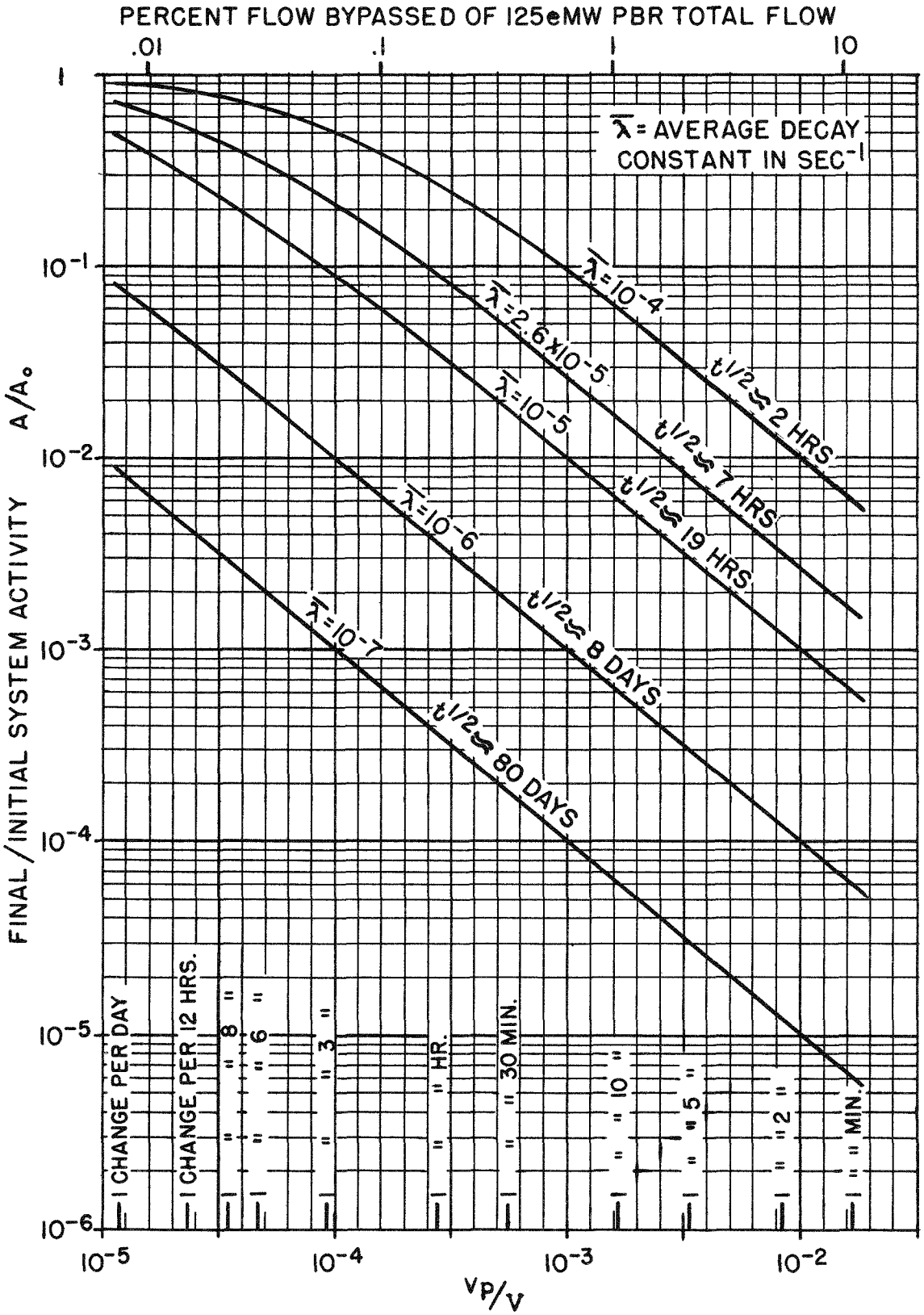
The development of coated particle fuel elements, capable of a high degree of fission product retention, can have a marked effect on the Commission gas cooled reactor program when it is recognized that the Pebble Bed Reactor is a high temperature gas cooled reactor capable of

using fissile and fertile material as an oxide and is a system that is relatively insensitive to dimensional changes in the moderator structure. In order to get a clear economic picture of the PBR potential, a design study should be made of a 300 eMW plant and costed for comparison with the fluid cooled nuclear plants recently studied by the Commission.



EFFECT OF ISOTOPE DECAY ON FISSION PRODUCT LEAKAGE RATE BASED ON 2-REGION COATED PARTICLE DIFFUSION MODEL

FIG. 1



EFFECT OF BYPASS CLEANUP ON SYSTEM ACTIVITY

FIG. 2

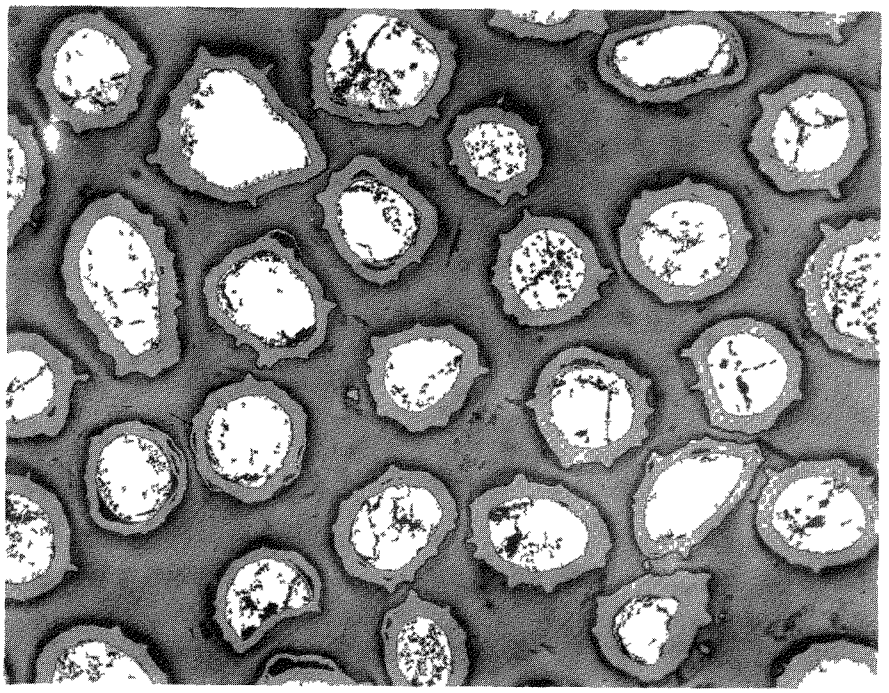


Figure 3. Vapor Deposited Al_2O_3 (20 micron) on Normal Enrichment UO_2 (105/149 micron). 100x

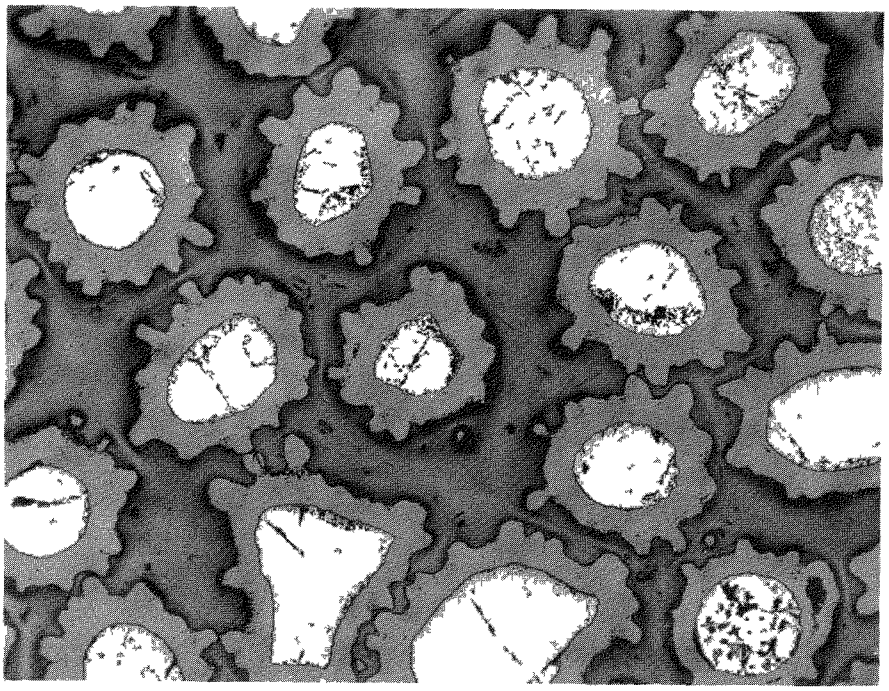


Figure 4. Vapor Deposited Al_2O_3 (38 micron) on Normal Enrichment UO_2 (105/149 micron). 100x

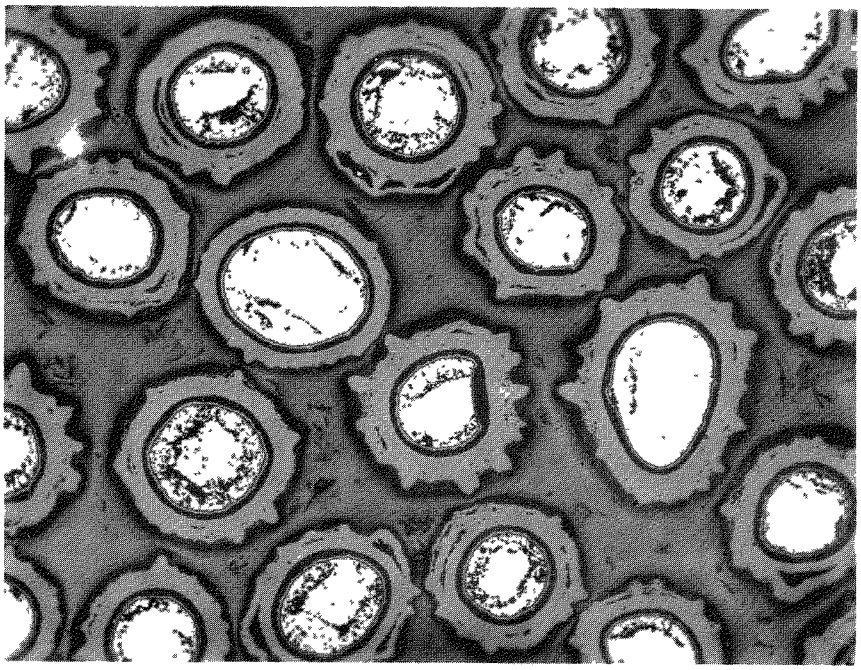


Figure 5. Vapor Deposited Al_2O_3 (55 micron) on Highly Enriched UO_2 (105/149 micron). 100x

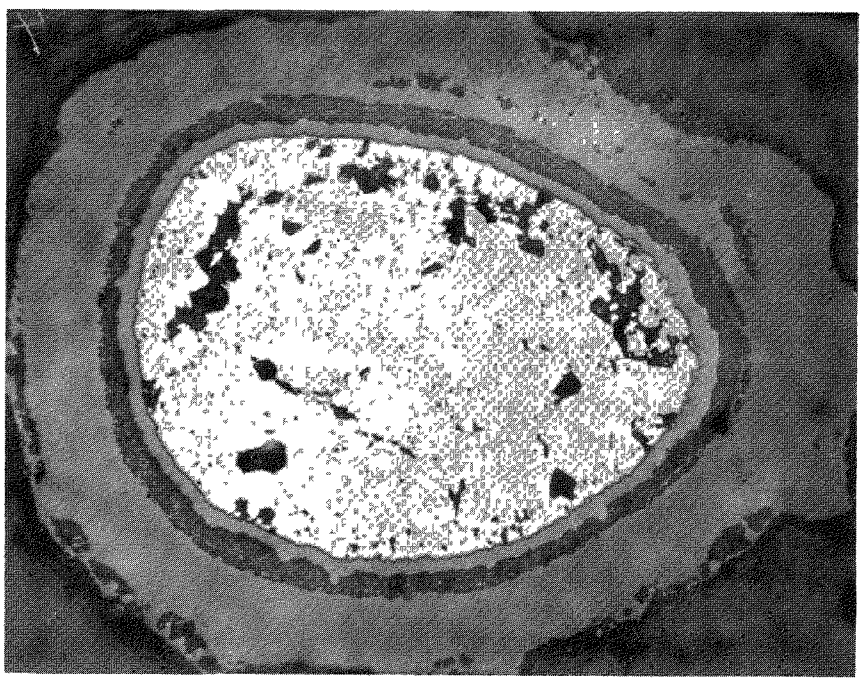


Figure 6. Vapor Deposited Al_2O_3 (55 micron) on Highly Enriched UO_2 (105/149 micron). 500x

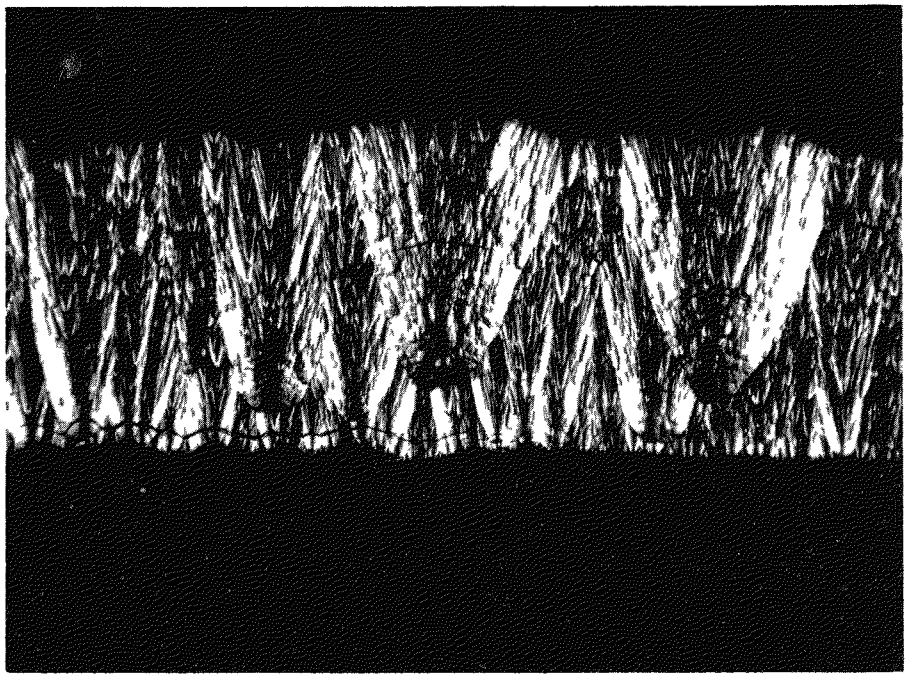
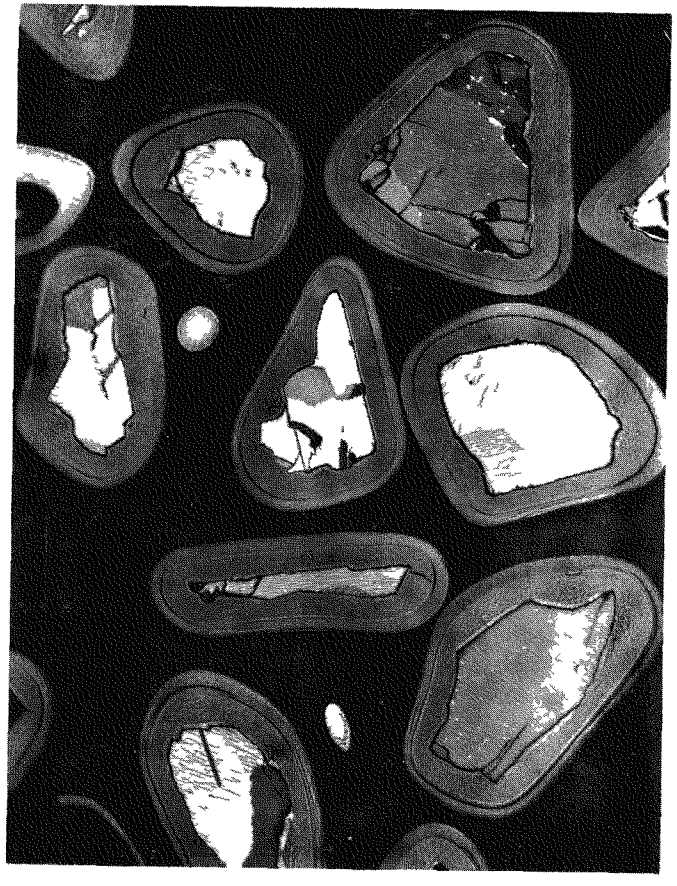
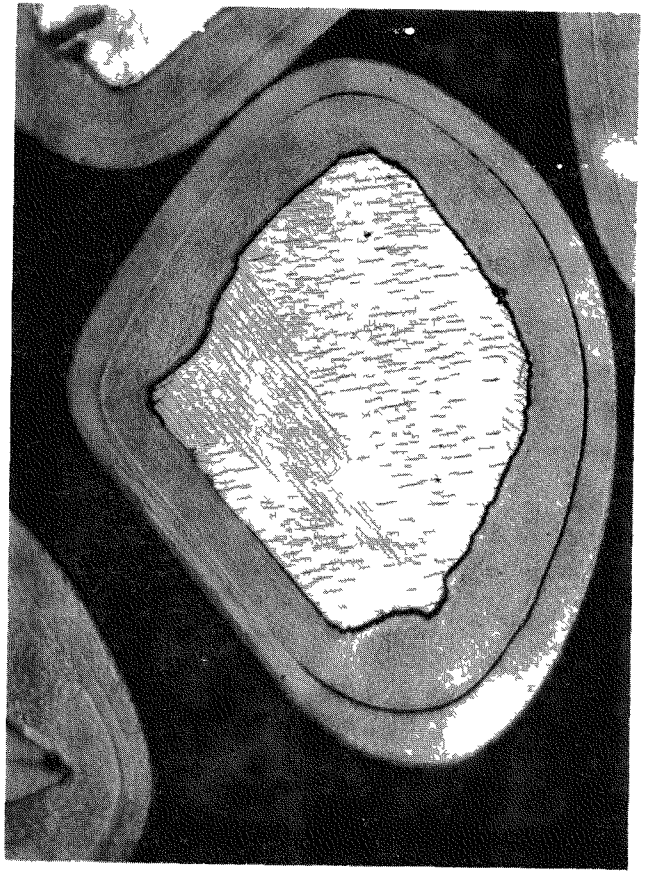


Figure 7. Pyrographite Coating at 250x (Raytheon)

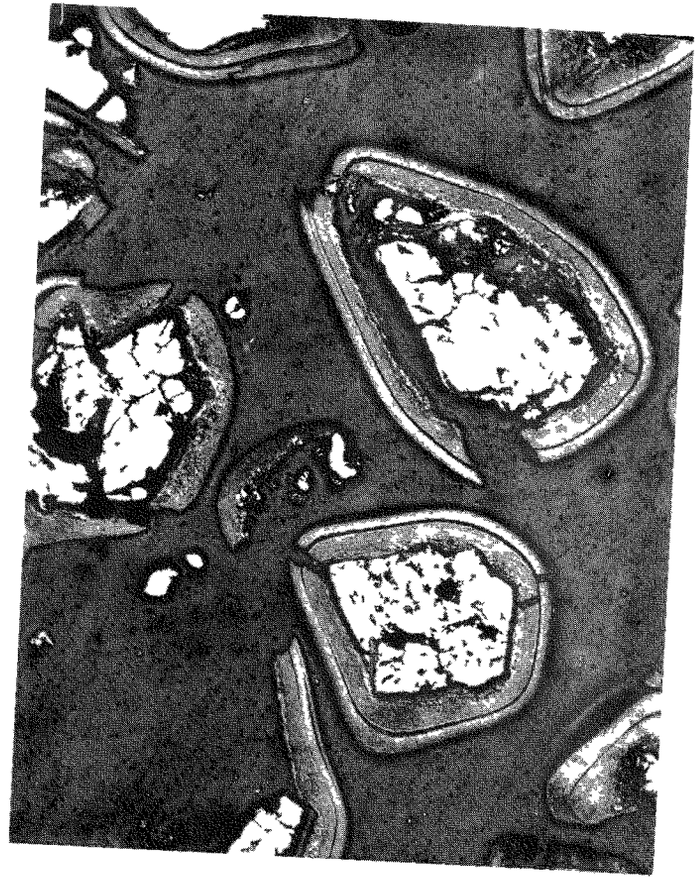


100x

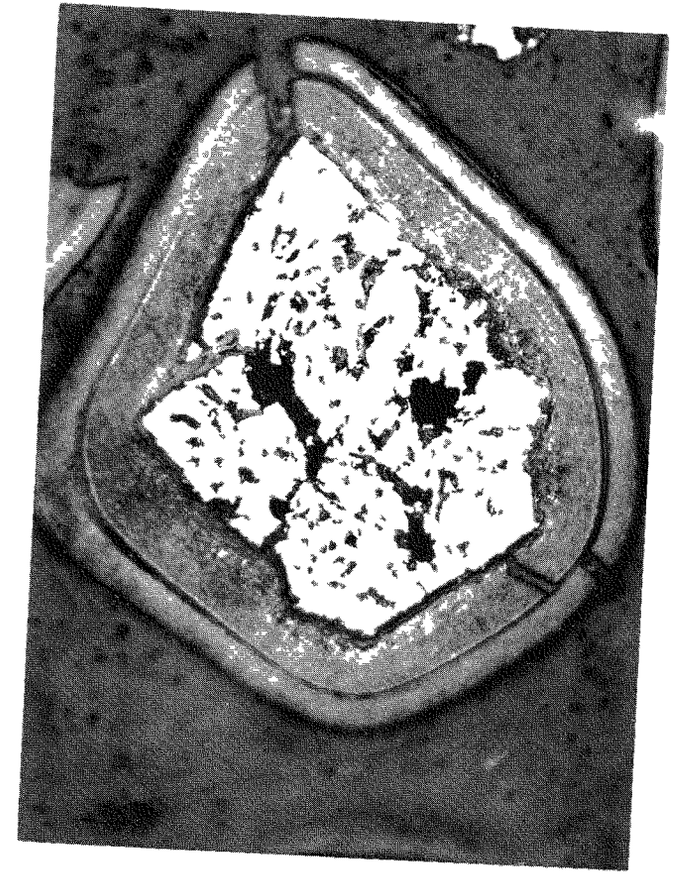


250x

Figure 8. Pyrolytically Deposited Carbon Coating on UC_2 Particles (BMI)



100x



250x

Figure 9. Pyrolytically Deposited Carbon Coating on UC_2 Particles After Thermal Cycling to 2000°C (BMI)