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WANL-TME-1329

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Westinghouse Astronuclear Laboratory

Contribution to the

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A. Fuel Particle Evaluation and Coating Development
(Title Unclassified)

by

A. L. Feild, Jr.

B. Experimental Investigation of Fission Product Release
(Title Unclassified)

by

G. L. Grandy

Classification cancelled (or changed to)
by authority of
by H.F.C. TIC, date SEP 11 1973

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INFORMATION CATEGORY

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A. Fuel Particle Evaluation and Coating Development
(Title Unclassified)

by

A. L. Feild, Jr.

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FUEL PARTICLE EVALUATION AND COATING DEVELOPMENT

1. EVALUATION OF VENDOR COATED PARTICLES

The evaluation of qualification, production, and development type coated particles from participating vendors has continued on a routine basis during the past six month period. Thermal migration testing has been conducted at 2300°C-4 hr. on all preproduction qualification and production lot samples, according to WANL-PDS-30050-B.

During this time, fuel particles have been furnished by General Atomic, 3M, and Carbon Products. All lots of fuel submitted by all three suppliers were acceptable from the standpoint of thermal behavior, with the portion of totally migrated particles averaging less than 1%. No difficulties were encountered in processing of these fuel particles into fabricated fuel elements.

2. COATING DEVELOPMENT USING METAL ADDITIVES IN PYROCARBON

At the last meeting of this group, the initial stages of a coating development program were described which had as an objective the co-deposition of PyC and a metal additive to form a more migration resistant particle coating. Aluminum and zirconium were selected as the most promising additive candidates for a number of reasons cited in the last meeting report. Sufficient experimental investigation has now been conducted to define more realistically the potential of this approach.

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In summary, the attempts to produce improved coatings with the addition of varying amounts of aluminum to the coating stream were unsuccessful. However, the use of zirconium, particularly in small amounts, appears to offer a definite enhancement of migration resistance in the duplex type of pyrocarbon coatings.

The work concerned with aluminum addition will be discussed first, followed by a discussion of results achieved with zirconium additive. As might be expected, the aluminum work was not as comprehensive since it was discovered early that the co-deposition of aluminum and PyC from the vapor phase could not be controlled in a satisfactory manner to produce the desired coating characteristics.

2.1 Coating Development with Aluminum Additive

A total of 17 experimental runs were made in the fluid bed coating apparatus using 10-g. charges of uncoated UC_2 particles. Aluminum, in the form of $AlCl_3$, was added in varying amounts to the CH_4 -He coating stream by means of a temperature controlled vaporizer. An inner PyC coating, of a laminar structure, was first deposited at $1200^\circ C$ to a thickness of 8-10 microns using a mixture of 97 v/o He-3 v/o CH_4 . No aluminum was used in this step. The outer coating, deposited at 1900° - $2000^\circ C$, consisted of a 93 v/o He-7 v/o CH_4 PyC coating stream, to which the desired quantity of $AlCl_3$ was added by He carrier flowing through the vaporizer before the stream entered the coating chamber. An outer coating thickness of 10-15 microns was desired. The PyC

coating parameters selected for both inner and outer coating were based on earlier run parameters which produced the best migration resistant laminar-granular combination coatings in the range of 20-25 microns thickness. The He flow rate was adjusted to give the optimum bed support and fluidizing conditions. The results of the experimental runs are detailed in Table I.

A typical photomicrograph of an as-coated particle (Run 1-B) is shown in Figure 1. A radiograph of the same material is shown in Figure 2. These pictures indicate the coating to be of irregular surface contour, and to be fluffy and generally porous. The vapor pressure of AlCl_3 in this run was 3mm. Subsequent runs, in which the vapor pressure of AlCl_3 ranged up to 640mm, showed a worsening of the coating structure. No graphitization of the PyC due to the fugitive presence of Al was detected, as shown in x-ray diffraction "d"-spacing analyses listed in Table I for all runs conducted. These "d" values are typical for a non-graphitic structure. In some cases, the inner coating, previously deposited without Al addition, seemed to be indistinguishable from the outer coating containing the Al.

A sample of particles from Run 1-B was heat treated at 2500°C for 1/2 hr. to determine uranium migration behavior. Conditions of radiographic exposure were 16Kv-5ma-3min. The particles performed poorly and showed complete uranium migration to the surface. No residual coating could be detected, as shown in Figure 3. Other runs, with higher aluminum additions during coating, also performed poorly.

Analysis of the overall results with aluminum as an additive indicated that it would be quite difficult to determine how, or whether, the coating process could be developed to produce desired conditions. Consequently, it was decided to concentrate on the alternate approach of zirconium additive and terminate the work on aluminum.

2.2 Coating Development with Zirconium Additive

During the last meeting, some initially encouraging results had been reported on a few preliminary coating runs utilizing zirconium as an additive in the outer PyC coating. This work has been continued and sufficient experimental data are available to show that the process does show promise for improving the migration resistance of UC_2 particles at temperatures around $2500^{\circ}C$.

In this procedure, Zr is added to the CH_4 -He coating stream in the form of $ZrCl_4$, which is supplied from the same type of controlled temperature vaporizer that was used with $AlCl_3$. Since both these materials are solid at temperatures of interest, the vapor pressure can be adjusted by temperature control to supply the amounts of salt required by sublimation. In the present work, $ZrCl_4$ pressures from 2-18 mm. were obtained over the temperature range 200° - $236^{\circ}C$. A purge of He through the vaporizer then transported the vapor into the main CH_4 -He coating stream.

A total of 44 experimental runs were made in which Zr was added to the outer PyC coating in varying amounts. The experimental data are shown in

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Table II. The inner pure PyC coating was initially deposited on the fluidized bed of uncoated UC₂ particles at either 1200°C or 1400°C, the latter temperature being somewhat more suitable. The helium carrier gas was diluted with 2.5-5.0 v/o CH₄ for this portion of the coating, and dense laminar structures of 5-10 microns thickness were produced. Following this, the temperature of the reaction chamber was raised to 1900°-2000°C, the vaporizer having been preheated to the desired operating temperature. With 2.5-8.0 v/o CH₄ in the He carrier, and the ZrCl₄ fed into the main coating stream, run times were adjusted to obtain approximately 12-16 micron thick doped outer coatings. The high temperature coating conditions for the PyC deposition were derived from optimum conditions of previous pure PyC coating runs aimed at production of dense, isotropic granular structures. The helium carrier flow in the bed was maintained at a value to provide a stable, well fluidized bed in the reaction chamber. As coating progressed, the flow requirement became less because of the decrease in composite density of the particles.

Evaluation of coatings produced was by means of chemical analysis, metallography, microradiography, x-ray diffraction, electron microscopy and electron microprobe analysis. Performance of coatings was determined by thermal migration testing at temperatures of 2300°C and 2500°C for times of 4 hrs. and 1/2-1 hr., respectively.

Chemical analysis for zirconium content in particles and coatings was performed using x-ray fluorescence, gravimetric analysis, and electron microprobe techniques. A number of microprobe traces across core and coating of

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Zr-containing samples indicated no Zr present in the core, and a very small amount in the inner coating, diffused from the interface between inner and outer coatings. Therefore, to relate Zr analyzed in the total particle (fluorescence and gravimetric) to Zr in the outer coating, an appropriate conversion factor was developed relating the assumed density and calculated volume of the outer coating to the composite density and volume of the entire particle. Generally, this concentration factor was in the range 2-10, depending on the relative coating thicknesses and particle size. This relationship is shown in the last two columns of Table II.

A series of photographs was prepared on two representative coating runs to illustrate the behavior of particles containing widely different Zr content in the coating. The runs selected were 10-A(0.20 w/o in particles) and 13-A(19.3 w/o in particles), which exhibited some differences in thermal performance.

The appearance of as-coated particles from these two runs is illustrated in Figures 4 and 5, in which polarized light was used. In both cases, the inner coating cannot be distinguished readily from the outer coating, due to migration of uranium into the inner coating during the high temperature outer coating deposition. Also, some inward diffusion of Zr into the inner coating during the outer coating deposition has occurred. The higher Zr content of Run 13-A is apparent from the presence of the white precipitate in the outer portion of the coating.

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Particles from Runs 10-A and 13-A heat treated at 2500°C for 1/2 hour are shown in the micrographs of Figures 6 and 7. In both cases the coatings have remained intact, but there is an increase in the amount of second phase in the coating which appears to be uniformly distributed.

An attempt to determine the structure of the second phase in these coatings was made by examination of the outer region of the coating using electron microscopy. The results of this study are shown in Figures 8-11, which were taken at magnifications of 10,000X. At low percentages of Zr (Run 10-A) the as-coated particles exhibit a scattered dispersion of second phase in the pyrocarbon matrix as shown in Figure 8. X-ray diffraction has identified this precipitate as ZrC. After exposure at 2500°C for 1/2 hour the precipitate is still present in a less apparent form, and the matrix structure has assumed a different texture (Figure 9). At high concentrations of Zr (Run 13-A) the structure appears more mottled and there are definite islands of ZrC imbedded in the matrix, illustrated in Figure 10. After a 2500°C-1/2 hour heat treatment, there are marked areas of ZrC agglomeration interspersed in a mixed background of ZrC and pyrocarbon (Figure 11).

A number of microradiographs were also made of particles containing low and high Zr contents, before and after heat treatment. Figure 12 shows particles of low Zr content (Run 10-A) radiographed in the as-coated condition. The difference in coating thickness seen in individual particles is due to the condition of the fluid bed operated with a small charge of particles.

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With only 10 gm. of particles in the bed, it is difficult to achieve uniform circulation of all particles in the bed, especially in a narrow particle size distribution. Consequently, some particles reside in the active coating area of the zone for longer times than others, leading to different amounts of coating deposited. Figure 12 indicates that no migration of uranium past the inner coating occurred during the coating operation. Radiographic exposure conditions were 7Kv-7ma-8min. As-coated particles from Run 13-A, containing high Zr content in the coating, were radiographed at 16Kv-5ma-3min. The additional power was required because of the absorption by the Zr in the coating. In order to delineate the core from the coating it is necessary to use sufficient x-ray power to eliminate the Zr-background while retaining the ability to detect uranium movement. Figure 13 shows this condition.

After heat treatment at 2500°C for 1/2 hour, radiography of particles with low Zr content (Run 10-A) indicates complete coating integrity, except for one particle, as shown in Figure 14. Again 16 Kv-5ma-3min. x-ray treatments were used, to delineate coating from fuel core. High Zr-content particles (Run 13-A) after the same heat treatment are seen in Figure 15. Again, one smaller particle appears to have suffered complete uranium migration, but the remainder exhibit an intact coating. At these higher levels of Zr, discernment of what is occurring through radiography becomes more difficult due to the higher net absorption of x-rays by the zirconium.

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On the basis of radiography, it can be concluded that a very high level of Zr content in the coating, such as in Run 13-A, offers no obvious advantage over low levels of Zr, as in Run 10-A, at least at temperatures of 2500°C for short times. Whatever mechanism is operative in the coating to reduce the migration of uranium, it apparently is effective with very low amounts of dispersed Zr.

Another set of radiographs were prepared to directly compare the performance in thermal testing of Zr-doped particles and a sample of representative commercial particles available, heat treated in the same crucible. Run 31-A was selected (1.42 w/o Zr in particles) from WANL material, and General Atomic lot 44001 served as the vendor material. The results of a 2300°C-4 hr. treatment are shown in Figure 16, where radiographic exposure conditions were 7Kv-7ma.-8min. Both types of particles survived this treatment without evidence of migration. Another thermal treatment of particles from the same lots at 2500°C for 1 hour resulted in the behavior shown in Figure 17. The WANL particles continue to maintain their integrity, and a considerable percentage of the vendor material appears unaffected. However, clumping of numbers of the latter particles, and exfoliation of others, indicates that gross migration has occurred in a substantial portion of the material.

Finally, two radiographs were made at reduced magnification (60X vs. normal 150X) to compare the performance of a large group of these two classes of particles under heat treatment at 2500°C for 1/2 hr. Results are illustrated in Figures 18 and 19. The vendor material (Figure 18) suffered gross migration

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of the majority of particles, with agglomeration occurring throughout the field of view. The WANL material with Zr doped coatings (Figure 19) showed no agglomeration, with each particle maintaining a separate identity. Some particles seem to have suffered local surface breakthrough, but the general performance is excellent under these exposure conditions.

3. CONCLUSIONS

The use of selected additives, co-deposited with a PyC coating on UC_2 fuel particles, appears to have the potential of raising by 100° - $200^\circ C$ the maximum exposure temperature of particles for times up to 1 hour. The work is of a preliminary nature, and was conducted in a small laboratory apparatus. Of necessity, scale-up factors must be investigated to determine applicability on a reasonable production basis. The laboratory effort has demonstrated that a closer degree of control and a more expensive coating operation will result from use of the additive technique, than from use of the conventional PyC coating methods.

From experimental evidence it appears doubtful that Al can be incorporated satisfactorily in PyC coatings by vapor deposition using the fluidized bed. However, Zr can be deposited in desirable quantities with a fairly reproducible behavior. Although a large number of runs were made (44) in the Zr investigation, more work on controlling the $ZrCl_4$ addition and a study of the kinetics of the system CH_4 - $ZrCl_4$ - H_2 are necessary.

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The results of WANL work on coating fuel particles have been transferred to all active fuel suppliers connected with the NERVA program. It is hoped that sufficient interest will be stimulated in these organizations to lead to more detailed laboratory investigations of the coating additive technique. Also, a summary report (WANL-TNR-208) has been prepared which describes in detail the entire fuel particle coating program conducted in the WANL laboratory.

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TABLE I

PyC + PyC and Al Coating Data

Run No.	Coating Temp. (°C)	Coating Time (min)	Gas Flow Rates-cc/min			CH ₄ V _f	Vaporizer Temp. (°C)	X-ray d (Å)
			He(Bed)	He(Sat)	CH ₄			
1-B	1200	90	2300	-	71	.03	-	3.44
	1900	30	1500	350	138	.07	122	
2-B	1200	90	2300	-	71	.03	-	3.44
	1900	45	1750	350	152	.07	120	
3-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	2100	350	181	.07	122	
4-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	2100	350	181	.07	121	
5-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	2100	300	181	.07	122	
6-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	2300	300	194	.07	122	
7-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	2300	300	194	.07	125	
8-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	2750	250	204	.07	122	
9-B	1200	90	2850	-	88	.03	-	3.44
	1900	30	2100	250	187	.07	125	
10-B	1200	90	2850	-	88	.03	-	3.43
	1900	30	1900	400	173	.07	144	
11-B	1200	90	2850	-	88	.03	-	-
			-	-	-	-	-	
12-B	1400	90	2500	-	77	.03	-	3.44
	1900	30	1900	450	181	.07	130	
13-B	1400	60	2500	-	77	.03	-	3.44
	1900	30	1900	450	181	.07	132	
14-B	1400	60	2500	-	77	.03	-	3.44
	1900	30	1900	225	166	.07	169	

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TABLE I (cont'd.)

Run No.	Coating Temp. (°C)	Coating Time (min)	Gas Flow Rates-cc/min			CH ₄ V _f	Vaporizer Temp. (°C)	X-ray d (Å)
			He(Bed)	He(Sat)	CH ₄			
15-B	1400	60	3000	-	93	.03	-	3.42
	1900	22	2500	200	203	.07	164	
16-B	1400	60	3000	-	93	.03	-	3.43
	2000	30	2500	-	183	.07	186	
17-B	1400	60	3000	-	93	.03	-	-
	2000	10	2300	-	173	.07	196	

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TABLE II

Duplex PyC+PyC and Zr Coating Data

Run No.	Coating Temp. (°C)	Coating Time (min)	Gas Flow Rates-cc/min			CH ₄ V _f (atm)	Sat. Temp. (°C)	ZrCl ₄ pp (mmHg)	w/o Zr	
			He(Bed)	He(Sat)	CH ₄				Particles	Outer Coating
1-A	1200	60	2500	-	67	.025	-	-	-	-
	1900	43	2150	500	58	.025	225	8.50	-	-
2-A	1200	30	1580	-	83	.05	-	-	-	-
	1900	43	1180	500	37	.03	224	8.00	-	-
3-A	1200	45	1580	-	83	.05	-	-	-	-
	1900	30	1075	500	33	.03	228	9.85	-	-
4-A	1200	60	1580	-	49	.03	-	-	-	-
	1900	22	1075	500	81	.07	220	6.50	-	-
5-A	1200	120	1800	-	56	.03	-	-	-	-
	2000	45	1180	500	89	.07	226	9.00	-	-
6-A	1200	60	1690	-	52	.03	-	-	-	-
	2000	90	785	500	59	.07	228	9.85	-	-
7-A	1200	45	3080	-	95	.03	-	-	-	-
	2000	60	1690	500	127	.07	236	18.50	-	-
8-A	1200	60	2600	-	80	.03	-	-	-	-
	2000	65	960	500	72	.07	225	8.50	12.10*	23.35
9-A	1320	45	2500	-	77	.03	-	-	-	-
	2000	50	880	500	66	.07	220	6.50	3.20*	-
10-A	1200	25	3300	225	102	.03	-	-	0.99**	3.85
	2000	22	3150	500	268	.07	203	2.65	0.199*	0.77
11-A	1200	66	3000	-	88	.03	-	-	2.65**	12.60
	2000	22	1500	550	158	.07	219	6.25	1.87*	8.92
12-A	1200	33	3000	-	88	.03	-	-	3.54**	15.72
	2000	31	1750	600	173	.07	220	6.50	2.51*	11.14
13-A	1200	58	3000	-	88	.03	-	-	19.94**	43.67
	2000	31	1650	650	173	.07	207	3.30	19.30*	42.27

* X-ray Fluorescence Analysis

** Gravimetric Analysis

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Run No.	Coating Temp. (°C)	Coating Time (min)	Gas Flow Rates-cc/min				Sat. Temp. (°C)	ZrCl ₄ pp (mmHg)	w/o Zr	
			He(Bed)	He(Sat)	CH ₄	CH ₄ V _f (atm)			Particles	Outer Coating
14-A	1200	11	1300	-	68	.05	-	-	1.30**	6.76
	2000	15	600	500	98	.08	219	6.25		
15-A	1200	14	1300	-	68	.05	-	-	3.57**	21.96
	2000	17	550	550	98	.08	223	7.70		
16-A	1200	14	1300	-	68	.05	-	-	2.37**	12.21
	2000	18	500	600	98	.08	226	9.00		
17-A	1200	11	1300	-	68	.05	-	-	1.82**	15.02
	2000	19	450	650	98	.08	225	8.50		
18-A	1200	11	1300	-	68	.05	-	-	2.10**	22.20
	2000	22	500	450	77	.07	222	7.30		
19-A	1200	14	1300	-	68	.05	-	-	1.21**	10.70
	2000	20	950	350	98	.07	220	6.50		
20-A***	1200	35	1500	-	45	.03	-	-	2.11**	16.00
	2000	25	900	400	98	.07	219	6.25		
21-A ⁺	1200	30	1900	-	59	.03	-	-		
	2050	25	1000	500	107	.07	130			
22-A	1200	60	2100	225	65	.03	-	-	2.10**	18.19
	2000	35	800	500	98	.07	207	3.30		
23-A	1200	60	2300	150	74	.03	-	-	2.95**	7.35
	2000	55	2100	400	188	.07	204	2.70	1.98*	4.93
24-A ⁺⁺	2000	30	600	400	76	.07	202	2.50	0.599*	
25-A	1200	60	2300	-	74	.03	-	-	1.92**	5.49
	2000	50	2300	150	190	.07	204	2.80		
26-A	1200	60	2300	-	74	.03	-	-	-	-
	2000	30	2000	500	190	.07	204	2.80		
27-A	1200	45	2300	-	74	.03	-	-	-	-
	2000	30	1900	500	190	.07	207	3.30		
28-A	1200	60	2400	-	74	.03	-	-	-	-
	1800	-	2300	450	197	.07	-	-		

*** 20g charge

+ AlCl₃ substituted for ZrCl₄

++ GA coated particles

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TABLE II (cont'd.)

Run No.	Coating Temp. (°C)	Coating Time (min)	Gas Flow Rates-cc/min			CH ₄ V _f (atm)	Sat. Temp. (°C)	ZrCl ₄ pp (mmHg)	w/o Zr	
			He(Bed)	He(Sat)	CH ₄				Particles	Outer Coating
29-A	1400	60	3000	-	93	.03	-	-	0.63*	2.04
	1900	30	2300	400	196	.07	203	2.65		
30-A	1400	40	3000	-	93	.03	-	-	0.96*	3.81
	1900	30	1900	400	173	.07	204	2.80		
31-A	1400	40	3000	-	93	.03	-	-	1.42*	6.28
	1900	30	1900	400	173	.07	210	3.85		
32-A	1400	25	3000	-	158	.05	-	-	0.49*	2.17
	1900	30	1900	400	173	.07	205	2.95		
33-A***	1400	40	2700	-	142	.05	-	-	0.26*	1.76
	1900	25	1500	400	113	.07	209	3.70		
34-A	1400	25	2500	-	132	.05	-	-	0.04*	1.58
	1900	30	1300	450	132	.07	185	-		
35-A	1400	25	2300	-	121	.05	-	-	0.62*	3.03
	1900	30	1100	450	113	.07	211	4.00		
36-A	1400	25	3000	-	158	.05	-	-	0.87*	4.72
	1900	30	900	450	98	.07	204	2.80		
37-A	1400	25	2300	-	121	.05	-	-	0.77*	4.00
	1900	30	900	500	113	.07	211	4.00		
38-A	1400	25	2300	-	121	.05	-	-	0.98*	4.82
	1900	30	1100	375	113	.07	212	4.30		
39-A	1400	25	2300	-	121	.05	-	-	0.64*	3.08
	1900	30	1100	400	113	.07	215	5.00		
40-A	1400	25	2300	-	121	.05	-	-	0.40*	1.99
	1900	30	1100	350	113	.07	209	3.70		
41-A	1400	25	2300	-	121	.05	-	-	1.28*	4.36
	1900	30	1000	350	113	.07	218	6.00		
42-A	1400	25	2300	-	121	.05	-	-	-	-
	1900	15	1000	425	113	.07	226	9.00		
43-A	1400	25	2300	-	121	.05	-	-	0.86*	6.08
	1900	13	1000	400	113	.07	200			
44-A	1400	25	3000	-	158	.05	-	-	0.72*	3.01
	1900	40	1000	450	109	.07	208	3.50		

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Figure 1. Micro Run 1-B As-coated (400X)

Figure 2. Radio Run 1-B As-coated (150X)
7Kv-7ma-8 min.

Figure 3. Radio Run 1-B 2500° - 1/2 hr. (150X)
16Kv-5ma-8 min.

Figure 4. Micro Run 10-A As-coated (400X)

Figure 5. Micro Run 13-A As-coated (400X)

Figure 6. Micro Run 10-A 2500° - 1/2 hr. (400X)

Figure 7. Micro Run 13-A 2500° - 1/2 hr. (400X)

Figure 8. E.M. Run 10-A As-coated (10,000X)

Figure 9. E.M. Run 10-A 2500° - 1/2 hr. (10,000X)

Figure 10. E.M. Run 13-A As-coated (10,000X)

Figure 11. E.M. Run 13-A 2500° - 1/2 hr. (10,000X)

Figure 12. Radio Run 10-A As-coated (150X)
7Kv-7ma-8 min.

Figure 13. Radio Run 13-A As-coated (150X)
16Kv-5ma-3 min.

Figure 14. Radio Run 10-A 2500° - 1/2 hr. (150X)
16Kv-5ma-3 min.

Figure 15. Radio Run 13-A 2500° - 1/2 hr. (150X)
16Kv-5ma-3 min.

Figure 16. Radio Run 31 + V.F. 2300°C - 4 hr. (150X)
7Kv-7ma-8 min.

Figure 17. Radio Run 31 + V.F. 2500°C - 1 hr. (150X)
7Kv-7ma-8 min.

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Figure 18. Radio V.F. 2500°C - 1/2 hr. (60X)
7Kv-7ma-8 min.

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Figure 19. Radio Run 31 2500°C - 1/2 hr. (60X)
16Kv-5ma-3 min.

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B. THE EXPERIMENTAL INVESTIGATION OF FISSION

PRODUCT RELEASE

(Title Unclassified)

G. L. GRANDY

INFORMATION CATEGORY
CONFIDENTIAL RESTRICTED DATA
John E. Sullivan 12/1/65
Authorized Classifier Date

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INTRODUCTION

The Fission Product Diffusion program is an experimental effort to determine the release of fission products as a function of time, temperature geometry, and environment from NERVA fuel.

The prime objectives of this program are:

- (1) To determine experimentally the magnitude of fission product release for times and temperatures characteristic of normal NERVA reactor operations.
- (2) To determine experimentally the magnitude of fission product release for times and temperatures which would be expected under accident conditions.
- (3) To incorporate the experimentally determined values into a computer program which can be used to predict fission product release for a NERVA reactor for under a variety of operating conditions.

TECHNIQUE

I Material

The fuel material used in these experiments consisted of nominally 100 micron diameter UC_2 spheres with a 25 micron layer of pyro-carbon coating. The coated spheres were distributed homogeniously in a graphite matrix at a concentration level of 20 percent by weight for

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Uranium. The U^{235} enrichment was approximately 93 percent. All samples were leached after preliminary machining to remove any exposed UC_2 . The fuel samples used to simulate accident conditions were heated to degrade the fuel particles and duplicates of each sample were photomicrographed to confirm the condition of the beads.

II Apparatus

All heating was done in a vacuum induction furnace which was capable of reaching pressures of $\sim 4 \times 10^{-4}$ torr. The temperature of the samples was measured with a dual wave length recording pyrometer.

III Calculation of Gross Release Fractions

The fractional release of gross fission product activity was determined by gamma counting the samples, before and after thermal treatment, in a constant geometry. The only correction required for this counting data was a decay correction for the time lapse between the first and second count. This decay correction was made empirically by counting an unheated "control" sample and using the single relationship:

$$A_C = A_1 C_2/C_1$$

where: A_C was the corrected activity in the test sample at time, t_2 .
 A_1 was the initial test sample activity at time, t_1 .

C_1 was the control sample activity at time, t_1 .

C_2 was the control sample activity at time, t_2 .

The fraction of the gross activity, f , retained in the test sample during the test was then given by:

$$f = A_t / A_C$$

where: A_t was the measured activity of the test sample at time, t_2 .

The release fraction was then simply $1 - f$.

For those samples on which measurements for individual nuclides were made the release fractions were measured radiochemically and suitable calculations applied.^(1,2,3)

IV Results

The fractional release of fission products as determined by this technique was used to compare two types of fuel material which were reported to have different UC_2 migration characteristics. The first type measured was material having a 25 micron pyrocarbon coat which was deposited in a single process. This material is referred to as type II fuel. The second material also had a 25 micron laminar coating but the pyrocarbon had been deposited in a two step process. This material is referred to as "duplex" fuel. Samples of both materials were heated for 5 minutes

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at various temperatures. The fractional release appeared to increase linearly with temperature and changed slope at $\sim 2250^{\circ}\text{C}$. The release data, as shown in Figure 1, appears to be the same for both types of material.

The difference in deposition techniques between the "duplex" and the type II fuel material studied result in essentially identically physical systems with respect to fission product diffusion. The diffusion path remains a 25 micron thickness of pyrocarbon.

Measurements were also made on the type II fuel material to determine release fractions for individual fission product nuclides. These studies were conducted at temperatures ranging from 1500 to 2300°C and time periods from 1 to 30 minutes. The nuclides investigated and the results are shown in Table I. Small release fractions of Sr^{89} and Ba^{140} were obtained at the lowest temperature studied (1500°C). This temperature appears to be a threshold for the release of fission products for NERVA fuel material with in the conditions utilized in this series.

No release of other nuclides was obtained until the fuel sample temperature was raised to 1900°C for ten minutes, at this point Ce^{144} and Y^{91} were not observed until a temperature of 2200°C was maintained for 30 minutes. No release of Zr^{95} and Ru^{103} occurred even at the most severe conditions studies studied, 2300°C for seven minutes.

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Release fractions for the various nuclides were found to be a complex function of time as can be seen in Figure 2 the logarithm of the fraction retained for Ba^{140} is linear, after an initial burst, at temperatures up to 2200°C . At 2200°C this same behavior was obtained over a 20 minute interval. After the 20 minute interval the release rate of Ba^{140} increased rapidly, giving essentially complete release of this nuclide in an additional 10 minutes. At 2300°C the release rates of Ba^{140} is linear throughout the time interval studied and is equivalent to the rapid rate obtained after 20 minutes at 2200°C . Sr^{89} behavior is similar to Ba^{140} under these conditions.

The release rates of other nuclides studied were linear with time and did not exhibit the initial burst effect. However, at 2200°C a drastic increase in release was again noted in samples heated longer than 20 minutes. The nuclides included in this group are Ce^{144} , Cs^{137} , Y^{91} , I^{131} , and Te^{129} .

EVALUATION OF DATA

The release data for Ba^{140} and Sr^{89} adhere to a double exponential diffusion model. These curves can be resolved into two exponential functions where the fraction retained is given by:

$$1 - f = f_1 e^{-D_1^i t} + (1 - f_1) e^{-D_2^i t}$$

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In this approach D'_1 and D'_2 are apparent diffusion constants with dimensions of reciprocal time. D'_2 may be evaluated by taking the slope of the linear portion of a $\log (1 - f_2)$ versus t plot. Extrapolation of this linear portion to $t = 0$, gives the value of $(1 - f_1)$ as an intercept. Values of D'_1 and f_1 may be obtained by substitution in the above equation.

The data for other nuclides gave diffusion constants which varied with time. For these nuclides another more or less empirical approach was used to evaluate the diffusion constants. For example Ce^{141} and Y^{91} release fractions were found to obey the first order rate equation:

$$1 - f = f_0 e^{-D'_1 t}$$

where: $1 - f$ = fraction retained
 f_0 = fraction of diffusing species present initially = 1
 D' = rate constant sec^{-1}
 T = time in seconds

Taking logarithms of both sides in the above equation one obtains:

$$\ln (1 - f) = \ln f_0 - D'_1 t$$

from which D' can be readily evaluated.

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EFFECT OF SAMPLE SIZE

The release data obtained to date have been measured on small samples. Since this data must be applied to fuel configuration for a NERVA reactor it was decided to investigate sample size effects. Cylindrical samples of beaded fuel material in a graphite matrix were prepared with diameters ranging from 1/16 to 1/4 inch. Neglecting end effects, these samples provided a change in diffusion path lengths of 1:2:4 and a volume ratio of 1:1.3:3 in ascending order of diameter.

In Table II release fractions are given for two temperatures, 1800°C and 2100°C. The only nuclide exhibiting size dependence was Ba¹⁴⁰, which decreased regularly with increasing sample size. All other nuclides were constant within experimental error and indicate that data from small samples can be extrapolated to other geometries.

FISSION PRODUCT RELEASE FROM TYPE III FUEL

It was decided that for the physical system of coated particle fuel in a graphite matrix, that diffusion through the pyrocarbon bead coat was the rate controlling step. To verify this conclusion samples of fuel were thermally treated to induce degradation of the bead coat prior to the fission product release experiments. The samples were heat treated in duplicate and photomicrographs of one sample were taken to verify coating

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degradation. This material was called type III fuel.

In almost all cases increased release was observed for the nuclides measured. Again no release was observed for Mo^{99} or Zr^{95} at the most severe conditions, 2300°C for 30 minutes. Conversely Ag^{111} , Sr^{89} , and Ba^{140} were almost completely released under the same conditions. The release fractions for Ba^{140} are shown in Figure 3. The marked increase in the initial burst can be seen for the type III fuel material.

The difference in gross fission product release between the two fuel types (II and III) is shown in Figure 4.

FIPDIF CODE

As was stated earlier one objective of this program is to incorporate the experimentally determined values for fission product release into a computer program. This program in turn can then be used to predict fission product release values for a reactor, such as NERVA, under a variety of operating conditions. To accomplish this task a modified (4) fission product inventory code, FIPDIF, has been written.

Several assumptions have been made in order to limit the experimental program to a reasonable size. Of the 499 isotopes which are applicable for such a program one can assume that all isotopes of a given element will diffuse at the same rate. While this assumption is not

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completely valid in an academic sense, it is sufficiently close to the actual situation to permit the prediction of the losses from a reactor core. Also, after a 10 minute power operation of a reactor such as NERVA, 96 percent of the activity produced in the fuel may be represented by only 21 elements individually contributing 0.7 percent or more of the total activity. A list of these elements and their relative contributions are given in Table III. These elements then comprise 142 isotopes and may be further classified into groups whose diffusion constants are of the same order of magnitude.

Using the above assumptions three cases have been run to check the accuracy of the FIPDIF code.

The first of these data came from an experiment that simulated a reactor loss of coolant accident using a $1\frac{1}{2}$ " long segment of a NERVA fuel element. The sample was irradiated then subjected to a post irradiation thermal treatment in a vacuum of $\sim 4 \times 10^{-4}$ torr. The sample was treated for one hour and the temperature varied from 2080°C to 2550°C as shown by the run profile in Figure 5. This run profile was predicted by another computer code, NOFLOW, for a 13 minute full power reactor operation followed by a complete loss of coolant. A FIPDIF program was then run using this temperature data and diffusion constant data obtained experimentally. The program predicted a gross reduction of 67.3 percent. The measured gross

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gamma reduction in the sample was 66.7 percent which is in remarkable agreement with the predicted value.

The other two cases used to check the FIPDIF program were actual reactor tests on the NRX-A2 and A3 reactors which were run at the Nuclear Rocket Development Station (NRDS) in Nevada.

The agreement between predicted and measured release values for these tests were not as close as those for the laboratory experiment as can be seen in Table IV. This discrepancy in release values can be accounted for in part due to two uncertainties. One, there is uncertainty in the measured release of fission products as determined by post mortem fuel analysis since the losses are small and the release values are based on differences in large numbers. Second, due to the complexity of the system it is difficult to reconstruct accurately a temperature profile for a complete reactor test. However, the prediction of releases from such tests within a factor of two is indicative that the technique is valid.

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FIGURE I

FRACTIONAL RELEASE OF GROSS GAMMA FISSION PRODUCT INVENTORY
FROM TYPE II AND DUPLEX NERVA FUELS HEATED 5 MINUTES IN VACUUM

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Time (Minutes)	Temp (Deg C.)	Sr ⁸⁹	Ba ¹⁴⁰	Cs ¹³⁷	Ce ¹⁴¹	Y ⁹¹	I ¹³¹	Te ¹²⁹	Zr ⁹⁵	Ru ¹⁰³
3	1500	0.032	0.008	0						
5	1500	0.068	0.003	0						
10	1500	0.145	0.051	0						
15	1500	0.072	0	0						
20	1500	0.114	0.011	0						
30	1500	0.182	0.064	0						
3	1600	0.044	0.019	0						
5	1600	0.069	0	0						
10	1600	0.121	0.010	0						
15	1600	0.126	0.004	0						
20	1600	0.162	0.009	0						
30	1600	0.159	0.035	0						
3	1800	0.177	0.052	0	0	0	0	-	0.021	-
5	1800	0.175	0.054	0	0	0	0	-	0.017	-
10	1800	0.175	0.050	0	0	0	0	-	0.00	-
15	1800	0.182	0.090	0	0	0	0	-	0.012	-
20	1800	0.188	0.104	0	0	0	0	-	0.00	0
30	1800	0.212	0.137	0	0	0.020	0	-	0.004	-
3	1900	0.202	0.075	0	0	0	0	-	0.004	-
5	1900	0.193	0.082	0	0	0	0	-	0.022	-
10	1900	0.238	0.128	0	0.007	0.020	0	-	0.021	-
15	1900	0.247	0.187	0	0.031	0.042	0	-	0.044	-
20	1900	0.163	0.138	0	-	-	0	-	-	0
30	1900	0.274	0.229	0	0.066	0.047	0	-	0.013	-
3	2000	0.242	0.134	0	0.000	0.009	0	-	0	-
5	2000	0.246	0.147	0	0.028	0.047	0	-	0	-
10	2000	0.275	0.191	0	0.042	0.011	0	-	0	-
15	2000	0.312	0.251	0	0.105	0.099	0	-	0	-
20	2000	0.228	0.145	0	0.013	0.053	-	-	0	0
30	2000	0.345	0.317	0	0.143	0.439	0	-	0	-
1	2200	0.168	0.104	0	-	0.061	-	-	-	-
3	2200	0.305	0.223	0	0.039	0.017	0	-	0	-
5	2200	0.302	0.217	0	0.046	0.078	0	-	0	-
10	2200	0.319	0.279	0	0.109	0.054	0	-	0	-
15	2200	0.347	0.304	0	0.129	0.143	0	-	0	-
20	2200	0.423	0.447	0.056	0.217	0.238	-	0.193	0	0
30	2200	0.999	0.999	0.905	0.949	0.800	0.358	-	0	-
3	2300	0.426	0.424	0.030	0.184	0.216	-	0.066	0	0
5	2300	0.839	0.846	0.199	0.500	0.339	-	0.001	0	0
7	2300	0.999	0.993	0.761	0.872	0.618	-	0.356	0	0

Table I Fractional Release of Fission Nuclides During Post Irradiation Thermal Anneals

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Figure 2 Diffusion of Ba¹⁴⁰ From UC₂ Graphite

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TABLE II

FRACTIONAL RELEASE OF NUCLIDES FROM CYLINDRICAL SAMPLES OF VARIOUS DIAMETERS

Sample Diameter (In Inches)	Time Min.	Temperature (°C)	Ba ¹⁴⁰	Sr ⁸⁹	Ce ¹⁴⁴	Y ⁹¹	Cs ¹³⁷
1/16	15	1800	0.105	0.184	--	--	0.019
1/8	15	1800	0.073	0.151	--	--	0.042
1/4	15	1800	0.070	0.163	--	--	0.030
1/16	15	2100	0.239	0.264	0.084	0.115	0.011
1/8	15	2100	0.220	0.248	0.064	0.089	0.043
1/4	15	2100	0.168	0.292	0.083	0.105	0.048

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¹⁴⁰Ba DIFFUSION IN TYPE III FUEL

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FIGURE 4
PERCENT OF FISSION NUCLIDES RELEASED FROM TYPES II AND III
FUEL AT 1800°C IN 30 MINUTES

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TABLE III

PERCENT CONTRIBUTION OF FISSION ELEMENTS TO TOTAL ACTIVITY
AFTER 10 MINUTE NERVA OPERATION

Element	%	Element	%
Rb	10.01	La	3.77
Nb	7.94	I	3.40
Sr	7.90	Sb	3.32
Y	7.89	Ce	3.21
Zr	6.70	Mo	3.10
Cs	6.57	Te	2.61
Kr	6.57	Se	2.19
Ba	5.10	Sn	1.25
Ru	5.14	As	0.72
Xe	4.81	Σ	96.02

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FIGURE 5

TEMPERATURE PROFILE FOR SIMULATED LOSS OF COOLANT ACCIDENT

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TABLE IV

COMPARISON OF GROSS FISSION PRODUCT RELEASE,
PREDICTED AND MEASURED

<u>TEST</u>	<u>PREDICTED</u>	<u>MEASURED</u>
Lab Expt.	67.3 %	66.7 %
NRX-A2	1.24%	1.02%
NRX-A3	1.6 %	0.9 %

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