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Detection and Control of As-Produced Pyrocarbon Permeability in Biso-Coated High-Temperature Gas-Cooled Reactor Fuel Particles

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DETECTION AND CONTROL OF AS-PRODUCED PYROCARBON PERMEABILITY IN BISO-COATED HIGH-TEMPERATURE GAS-COOLED REACTOR FUEL PARTICLES

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D. P. Stinton, B. A. Thiele,* W. J. Lackey,
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ABSTRACT

Fuel for High-Temperature Gas-Cooled Reactors consists of dense uranium carbide and thoria microspheres coated with layers of pyrolytic carbon and silicon carbide. The pyrolytic carbon coatings must be impermeable to fission gases to function properly during irradiation. Therefore, particles must be carefully characterized to determine as-produced permeability. The experiment described here compares several methods of permeability measurement and evaluates their value in predicting irradiation stability.

About 60 Biso-coated particle batches with coatings deposited in either 0.13- or 0.24-m-diam coaters were studied in this work. These batches were carefully characterized for permeability by neon-helium intrusion, long-term chlorination followed by radiography, and fission gas release. These methods of permeability measurement were compared and correlated with deposition conditions as well as pyrocarbon properties. The results from several irradiation tests were also used to evaluate the validity of the permeability measurements.

The neon-helium and long-term chlorination techniques correlated very clearly. Coatings with neon-to-helium ratios below 0.3 were gastight by the chlorination procedure, whereas those with ratios above 0.4 were permeable. The fission gas release technique was unable to distinguish between slightly permeable coatings and gastight ones. Therefore, neon-helium and long-term chlorination procedures are preferred over the fission gas release technique. Results from several irradiation tests verified that coatings with neon-to-helium ratios below 0.3 were gastight, whereas those with ratios above about 0.4 were permeable.

INTRODUCTION

Coated microspheres less than 1 mm in diameter have been developed as fuel for High-Temperature Gas-Cooled Reactors (HTGRs). Fertile

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*KFA, Jülich, West Germany.

microspheres are coated with two layers of pyrolytic carbon (PyC) and are called Biso particles. The denser outer PyC layer serves as the diffusion barrier to gaseous fission products.[1] In addition to an initial porous carbon layer, fissile microspheres are coated with a 35-μm layer of silicon carbide (SiC) sandwiched between two PyC layers and are called Triso particles. These layers provide the diffusion barrier to solid fission products.[1] Polished metallographic cross sections of typical fertile and fissile particles are shown in Fig. 1.

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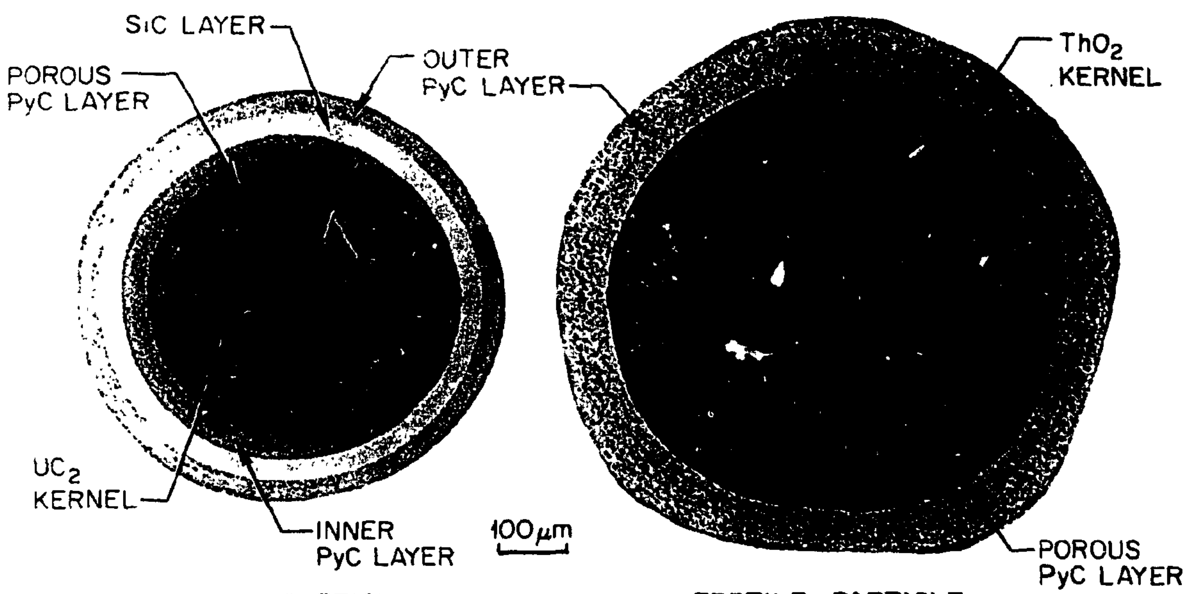

Fig. 1. Reference High-Temperature Gas-Cooled Reactor Fuel Particles.

In both fissile and fertile particles the pyrocarbon layers must be impermeable. For a Biso particle the coatings have failed when they become permeable and release gaseous fission products. In the past it was believed that the inner PyC layer of a Triso-coated particle [inner low-temperature isotropic (LTI)] was only a structural layer that had to withstand the pressures generated by fission products and protect the SiC layer from chemical attack.[2] Recent postirradiation examinations indicated that inner LTI layers must be impermeable to chlorine that is present during deposition of the SiC layer.[3,4] Chlorine is produced during the thermal decomposition of methyltrichlorosilane (CH_3SiCl_3), which forms the SiC layer. If chlorine penetrates a permeable PyC layer during SiC deposition, chlorine will react with rare earth fission products generated during the irradiation. These highly volatile rare earth chlorides diffuse through the PyC layer and cause severe corrosion and possible failure of the SiC layer.

Our study includes a description of several test methods used to determine whether as-produced pyrocarbon coatings were gastight or permeable. About 60 Biso-coated ThO_2 particle batches (Tables 1 and 2) were carefully examined by these techniques to determine the pyrocarbon permeability. The as-produced permeability was then correlated with properties of the pyrocarbon layers and with process variables used to deposit the coatings.

EXPERIMENTAL PROCEDURE

During the past several years the chlorination, neon-helium instrusion, and fission gas release techniques have been developed to determine whether preirradiated pyrocarbon coatings were gastight or permeable. One technique that has been described previously[5] in detail uses a long-term chlorination followed by a careful examination by microradiography. This technique is a modification of a chlorine leach procedure[6] used to detect cracked or missing pyrocarbon coatings. In this technique particles are exposed to chlorine at 1500°C for 18 h. During the treatment at 1500°C, chlorine gas can diffuse through the pyrocarbon layer of permeable coatings and react with the heavy metal of the kernel. The heavy metal chlorides diffuse into the buffer and, depending upon the degree of pyrocarbon permeability, also diffuse through the LTI layer. If heavy metal chlorides are present in the buffer layer, the LTI coating must be permeable to chlorine. Thorium tetrachloride ($ThCl_4$) can be detected in the buffer layer by x-ray microradiography after the particles have been chlorinated (Fig. 2). Many batches of particles, each in a separate container, can be chlorinated simultaneously since only the radiographs must be examined.

Table 1. Coating Parameters of the Biso-Coated ThO_2 Particle Batches

Run	Hydro-carbon[a]	Diluent		Charge Size (kg)	LTI		Immersion Density, Mg/m^3		Open Porosity (%)	Coating Thickness (μm)
		Type	(%)		Deposition Temperature (°C)	Coating Rate (μm/min)	Regular	Corrected for Open Porosity		
A-747	P	He	50	2.1	1225	1.17	1.948	1.860	4.51	29
A-754	P	H_2	67	2.1	1250	0.35	1.955	1.875	4.10	32
A-758	M	He	67	2.1	1350	4.92	1.755	1.664	5.24	81
A-762	M	He	67	2.1	1225	3.25	1.962	1.904	2.96	71
A-764	P	He	67	2.1	1350	3.29	1.864	1.811	2.82	92
A-765	P	Ar	50	2.1	1350	3.14	1.915	1.858	2.99	79
A-766	M	H_2	50	2.1	1350	4.78	1.764	1.672	5.20	66
A-768	P	H_2	50	2.1	1225	0.61	2.005	1.957	2.39	44
A-770	P	Ar	67	2.1	1225	1.25	2.012	1.992	1.00	94
A-771	M	H_2	67	2.1	1350	4.18	1.698	1.619	4.63	69
A-772	M	Ar	67	2.1	1350	4.41	1.693	1.593	5.94	73
A-775	M	Ar	50	2.1	1350	4.79	1.729	1.558	9.90	79
A-779	P	Ar	50	2.1	1225	2.10	2.019	1.992	1.32	126
A-780	P	He	50	2.1	1350	4.03	1.864	1.773	4.92	78
A-782	P	H_2	50	2.1	1350	2.95	1.855	1.810	2.43	88
A-783	M	He	50	2.1	1350	5.95	1.732	1.530	11.70	86
A-784	M	He	50	2.1	1225	4.44	1.929	1.842	4.52	89
A-785	M	Ar	67	2.1	1225	3.05	1.872	1.815	3.02	76
A-786	M	Ar	50	2.1	1225	3.67	1.897	1.826	3.75	88
A-787	P	H_2	67	2.1	1350	2.68	1.738	1.691	2.71	81
A-803	P	Ar	67	2.1	1350	2.46	1.798	1.728	3.92	71
A-804	P	He	67	2.1	1225	2.25	1.987	1.931	2.82	70
A-806	M	H_2	67	2.1	1225	2.82	1.943	1.878	3.33	71
A-807	M	H_2	50	2.1	1225	3.63	1.921	1.867	2.82	98
J-635	M	Ar	67	2.1	1250	4.41	1.925	1.862	6.21	71
J-636	M	Ar	67	10.0	1150	2.97	1.938	1.885	2.70	79
J-637	P	Ar	50	10.0	1375	4.13	1.918	1.837	4.21	92
J-638	M	Ar	67	10.0	1250	4.13	1.945	1.862	4.26	76
J-639	P	Ar	50	10.0	1325	3.36	2.001	1.941	3.03	90
J-640	M	Ar	67	12.5	1200	3.75	1.950	1.867	4.23	81
J-641	M	Ar	67	10.0	1200	3.27	1.951	1.881	3.58	73
J-642	M	Ar	67	15.0	1250	3.15	1.934	1.817	6.03	71
J-643	P	Ar	50	10.0	1325	3.52	1.989	1.926	3.20	74
J-644	M	Ar	67	12.5	1250	4.37	1.933	1.751	9.42	85
J-645	M	Ar	67	10.0	1200	3.76	1.941	1.870	3.62	77
J-646	M	Ar	67	15.0	1250	4.17	1.943	1.745	10.18	77
J-647	P	Ar	50	10.0	1425	5.08	1.958	1.850	5.52	76
J-648	M	Ar	67	15.0	1200	3.42	1.945	1.816	6.62	73
J-649	M	Ar	67	10.0	1250	3.57	1.939	1.863	3.57	75
J-650	P	Ar	50	10.0	1350	3.32	1.993	1.925	3.38	74
J-651	P	Ar	50	10.0	1425	4.38	1.903	1.799	5.47	77
J-652	M	Ar	67	10.0	1225	4.03	1.942	1.862	4.15	85
J-653	M	Ar	67	10.0	1150	2.76	1.951	1.901	2.56	83
J-654	M	Ar	67	10.0	1250	3.62	1.920	1.838	4.27	80
J-655	P	Ar	50	10.0	1375	4.11	1.987	1.914	3.70	80
J-835	P	Ar	50	10.0	1375	4.07	1.799	1.645	8.56	86
J-836	P	Ar	50	10.0	1375	4.42	1.780	1.627	8.55	82
J-838	M	Ar	50	10.0	1225	3.24	1.855	1.712	7.73	88
J-839	M	H_2	50	10.0	1200	4.14	1.763	1.561	11.42	93
J-840	P	Ar	50	10.0	1325	3.40	1.916	1.829	4.51	78
J-841	P	Ar	50	10.0	1325	3.76	1.924	1.840	4.37	73
J-842	M	Ar	50	10.0	1200	3.94	1.908	1.760	7.75	75
J-843	M	Ar	50	10.0	1250	2.65	1.835	1.682	8.32	89
J-844	M	Ar	50	12.0	1225	3.33	1.879	1.725	8.19	80
J-845	M	Ar	50	12.0	1250	3.00	1.858	1.659	10.70	89
J-846	P	H_2	50	10.0	1375	4.30	1.803	1.602	11.14	79
J-847	P	Ar	50	12.0	1325	3.78	1.937	1.857	4.12	91
J-848	P	H_2	50	10.0	1325	3.64	1.926	1.841	4.44	77
J-849	M	Ar	50	10.0	1200	4.44	1.925	1.720	10.65	83
J-850	P	H_2	50	12.0	1325	3.27	1.823	1.743	4.41	77

[a]P = propylene; M = mixed gas.

Table 2. Coating Properties of the Biso-Coated ThO_2 Particle Batches

Run	Fraction of Defective Particles			Ne/He	BAF_o[a]
	2-h Chlorine Leach	Fission Gas Release	Long-Term Chlorination and Radiography		
A-747	1.9×10^{-3}	2×10^{-5}	Gastight	0.15	1.474
A-754	1.1×10^{-3}	9×10^{-6}	Gastight	0.21	1.226
A-758	1.0×10^{-4}	5×10^{-6}	Permeable	0.48	1.027
A-762	1.3×10^{-5}	$<1 \times 10^{-6}$	Gastight	0.36	1.043
A-764	4.9×10^{-6}	$<1 \times 10^{-6}$	Slightly permeable	0.35	1.033
A-765	1.8×10^{-5}	$<2 \times 10^{-6}$	Slightly permeable	0.34	1.035
A-766	8.3×10^{-6}	$<2 \times 10^{-6}$	Permeable	0.51	1.024
A-768	2.1×10^{-3}	2×10^{-6}	Gastight	0.13	1.343
A-770	5.7×10^{-4}	3×10^{-6}	Gastight	0.15	1.107
A-771	5.0×10^{-6}		Permeable	0.44	1.030
A-772	2.0×10^{-5}		Permeable	0.66	1.031
A-775	6.4×10^{-4}	7×10^{-3}	Permeable	0.70	1.030
A-779	5.7×10^{-5}		Gastight	0.19	1.055
A-780	4.2×10^{-6}	$<2 \times 10^{-6}$	Permeable	0.37	1.022
A-782	5.2×10^{-5}	$<1 \times 10^{-6}$	Slightly permeable	0.27	1.026
A-783	7.4×10^{-4}	$<2 \times 10^{-6}$	Permeable	0.60	1.023
A-784	1.5×10^{-6}	$<3 \times 10^{-6}$	Permeable	0.43	1.029
A-785	8.6×10^{-7}	$<1 \times 10^{-6}$	Permeable	0.32	1.025
A-786	8.9×10^{-7}	$<2 \times 10^{-6}$	Permeable	0.36	1.030
A-787	1.5×10^{-5}	4×10^{-6}	Slightly permeable	0.35	1.031
A-803	4.2×10^{-5}	$<4 \times 10^{-6}$	Permeable	0.43	1.037
A-804	4.4×10^{-5}		Gastight	0.27	1.055
A-806	1.1×10^{-6}	$<2 \times 10^{-6}$	Gastight	0.38	1.036
A-807	5.8×10^{-5}	$<2 \times 10^{-6}$	Slightly permeable	0.28	1.032
J-635	1.0×10^{-3}		Permeable	0.62	1.033
J-636	2.3×10^{-3}		Gastight	0.29	1.059
J-637	3.6×10^{-6}		Permeable	0.70	1.033
J-638	1.1×10^{-4}		Permeable	0.26	1.049
J-639	8.0×10^{-5}		Gastight	0.18	1.046
J-640	1.0×10^{-3}		Permeable	0.39	1.054
J-641	1.7×10^{-4}		Slightly permeable	0.30	1.049
J-642	1.0×10^{-4}		Permeable	0.69	1.048
J-643	8.6×10^{-4}		Gastight	0.18	1.057
J-644	3.6×10^{-4}		Permeable	0.70	1.040
J-645	1.3×10^{-3}		Slightly permeable	0.35	1.045
J-646	2.5×10^{-4}		Permeable	0.70	1.048
J-647	6.5×10^{-6}		Permeable	0.46	1.043
J-648	2.1×10^{-4}		Permeable	0.68	1.054
J-649	1.2×10^{-4}		Permeable	0.36	1.042
J-650	3.5×10^{-4}		Permeable	0.55	1.045
J-651	4.4×10^{-5}		Permeable	0.42	1.031
J-652	6.3×10^{-6}		Permeable	0.27	
J-653	1.2×10^{-3}		Gastight	0.24	1.060
J-654	4.8×10^{-6}		Permeable	0.39	
J-655	7.1×10^{-5}		Slightly permeable	0.22	1.047
J-835	8.5×10^{-5}		Permeable	0.52	1.028
J-836	3.5×10^{-4}		Permeable	0.39	1.028
J-838	1.1×10^{-4}		Permeable	0.63	1.029
J-839	3.0×10^{-3}		Permeable	0.48	1.022
J-840	3.0×10^{-5}		Gastight	0.29	1.037
J-841	1.9×10^{-4}		Gastight	0.30	1.039
J-842	7.7×10^{-5}		Permeable	0.61	1.030
J-843	2.6×10^{-5}		Permeable	0.48	1.028
J-844	3.9×10^{-4}		Permeable	0.60	1.032
J-845	2.2×10^{-3}		Permeable	0.56	1.031
J-846	5.5×10^{-3}		Permeable	0.54	1.023
J-847	2.1×10^{-5}		Gastight	0.27	1.041
J-848	8.0×10^{-5}		Gastight	0.26	1.032
J-849	4.2×10^{-4}		Permeable	0.57	1.029
J-850	2.5×10^{-2}		Permeable		1.034

[a]Bacon Anisotropy Factor determined optically.

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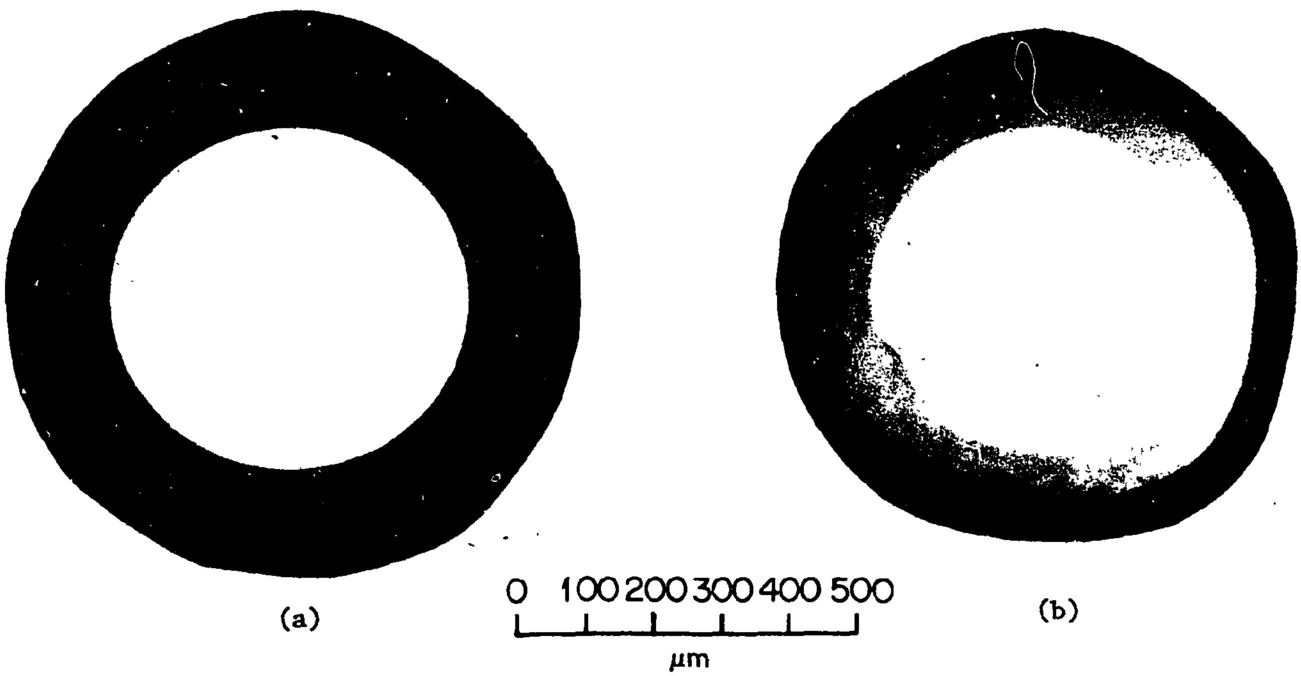

Fig. 2. Typical X-Ray Microradiograph of (a) a Particle with a Gastight PyC Coating and (b) a Particle with a Gas-Permeable PyC Coating and Heavy Metal Migration into the Buffer Layer. Microradiographs were made after an 18-h chlorination at 1500°C.

A second method used to characterize pyrocarbon coatings for permeability is the neon-helium intrusion technique.[7] Particles are annealed at 1375°C in an atmosphere of half neon and half helium for 1 h. During this time individual neon and helium atoms diffuse through the passages in the LTI layer and into the buffer layer. Since the helium atoms are very small, they diffuse through the LTI layer and saturate the buffer with helium. Neon is a larger atom and does not pass as easily through the passages to the buffer. Therefore, neon only partially fills the voids in the buffer layer. Helium and neon release are monitored as particles are individually broken in a vacuum chamber connected to a mass spectrograph. The ratio of the volume of neon to the volume of helium is a measure of the coating's permeability to neon. Low values indicate gastight coatings since neon diffused through the pyrocarbon layer much slower than helium. A high ratio indicates a coating permeable to neon.

The final method used to characterize coated paricles for permeability is fission gas release.[8] In this technique the particle samples of interest are bombarded by a high-intensity beam of photons generated in a linear accelerator. The impinging beam of x-ray photons induces fissioning of the thorium fuel, creating a noble fission gas inventory. The fission gases given off are then collected for gamma counting and analysis. By comparison with a concurrently irradiated standard, release-to-birth (R/B) ratios for the fission gases of interest are calculated.

RESULTS

Coated particles must remain impermeable to fission gases to perform adequately during irradiation. To produce gastight coatings the characteristics that affect coating permeability must be determined. Studies were performed to carefully examine the following coating characteristics, which were thought to control coating permeability: thickness, anisotropy, open porosity, and density. The desired LTI thickness was 75 μm for all the developmental coating runs. Because of the many variables involved during coating deposition, the coating thickness typically varied from 65 to 95 μm. Within this range of thicknesses no correlation existed between coating thickness and the coating permeability as determined by either chlorine leach or neon-helium intrusion (Fig. 3). Obviously, where extremes of thickness are involved, an effect on permeability would be expected.[9] Apparently coatings with thicknesses within the range mentioned above are either permeable or gastight because of their structure and not simply because of coating thickness.

Coatings must be nearly isotropic to survive stresses generated during irradiation. Anisotropy was measured by using the Bacon Anisotropy Factor determined optically (BAF_o).[10-12] The BAF_o values were measured by General Atomic Company personnel with their "Seibersdorf" equipment. Values below 1.05 are thought to be sufficiently isotropic to survive irradiation. In the range $BAF_o < 1.05$, permeability seemed to be independent of the BAF_o value (Fig. 4). That is, for coatings with BAF_o values below 1.05, half were permeable and half were gastight. Certainly as the BAF_o values

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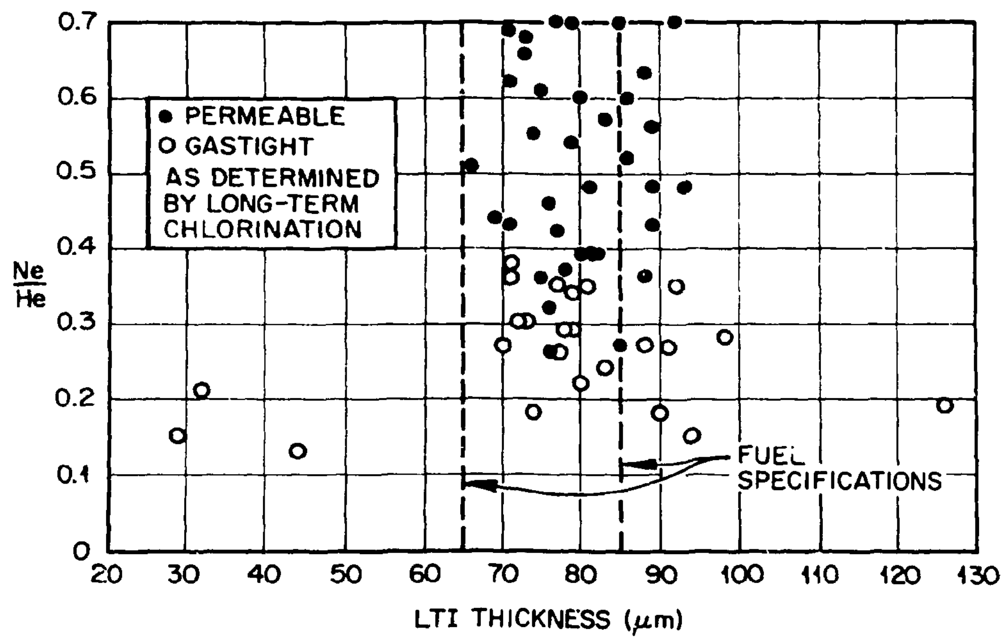

Fig. 3. Coating Thickness Had Little Effect on Coating Permeability.

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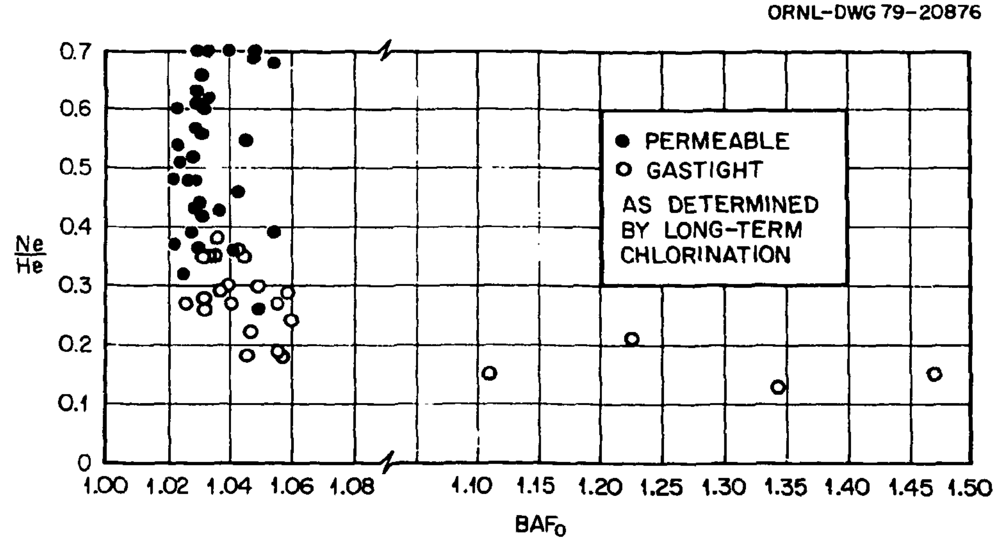

Fig. 4. At Values of the Bacon Anisotropy Factor Determined Optically (BAF_O) Less than 1.05, the Permeability of the Coating Could Not Be Correlated with the BAF_O Value.

increased to very high values, the coatings became gastight. In Fig. 4 coatings were determined to be permeable by using the chlorine leach and radiograph technique.

The final two properties, percent open porosity and bulk density, had significant influence on coating permeability. The open porosity[13] of a coating was determined from mercury porosimetry measurements and the volume of the LTI as determined from a radiograph. Figure 5 shows that, as the percent of open porosity increases, the neon-to-helium ratios increase, indicating that the coatings are becoming more permeable. No clear boundary at one specific porosity level determines whether coatings are permeable or not; however, the trend is clear.

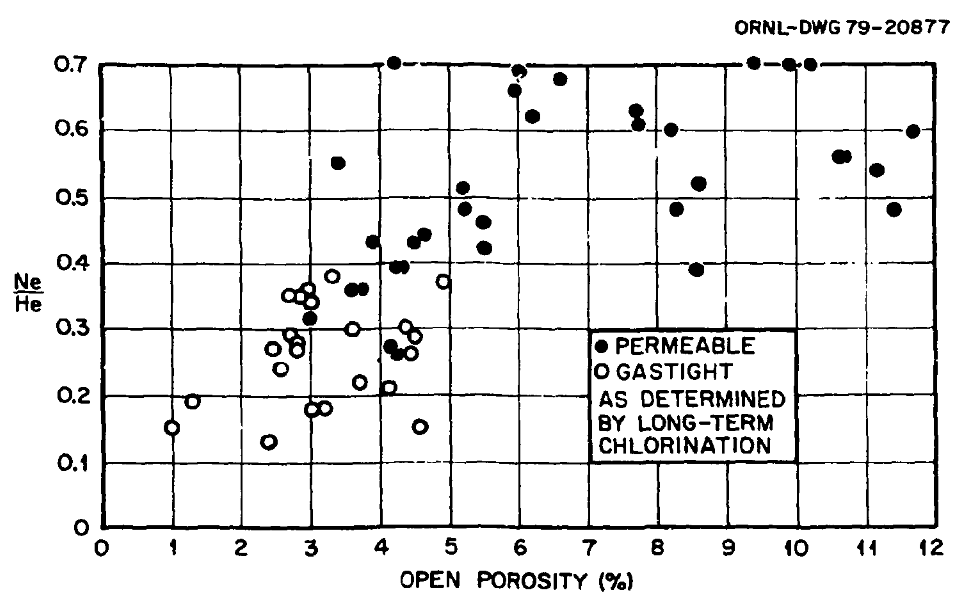

Fig. 5. Large Amounts of Open Porosity Cause Pyrocarbon Coatings to Become Permeable.

The second coating characteristic that influenced the permeability was the corrected immersion density.[13] This is a measure of the bulk density of the LTI coating found by obtaining an immersion density of LTI fragments and correcting it for the amount of open porosity present. Figure 6 shows that, as the density decreases, the coatings tend to become more permeable (higher neon-to-helium ratios). Again, there is no clear cutoff, but a definite trend is apparent.

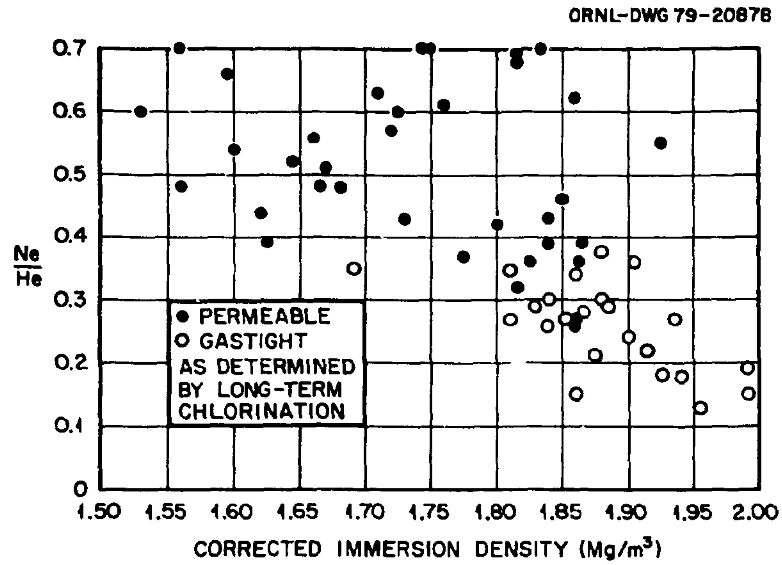

Fig. 6. Coatings with High Densities Are More Likely to Be Gastight.

Several process variables must be controlled to produce gastight LTI coatings, that is, ones with small amounts of open porosity and high corrected gradient densities. One of the most important variables is the temperature at which pyrocarbon coatings are deposited. Deposition temperature is a major variable in controlling the density of the coating.[14] High temperatures produce coatings that have low densities and are more likely to be gas permeable. Figure 7 shows the results from numerous coatings deposited with a mixture of propylene and acetylene in a 0.24-m-diam coater. All the coatings deposited at 1350°C had low densities and were permeable as determined by the chlorine leach and radiograph procedure. Coatings deposited at temperatures between 1150 and 1250°C had higher densities and were usually gastight. Batch size also had a marked effect on coating permeability, as shown in Fig. 7. The normal batches in this figure used 10 kg of kernels. When the batch size was increased to 12.5 or 15.0 kg and the process variables were held constant, the properties of the pyrocarbon changed and the coatings became permeable.

The coatings examined in this study were deposited in both 0.13- and 0.24-m-diam coaters. The gas distribution system must uniformly distribute hydrocarbon to prevent low-density inclusions within coatings, which cause

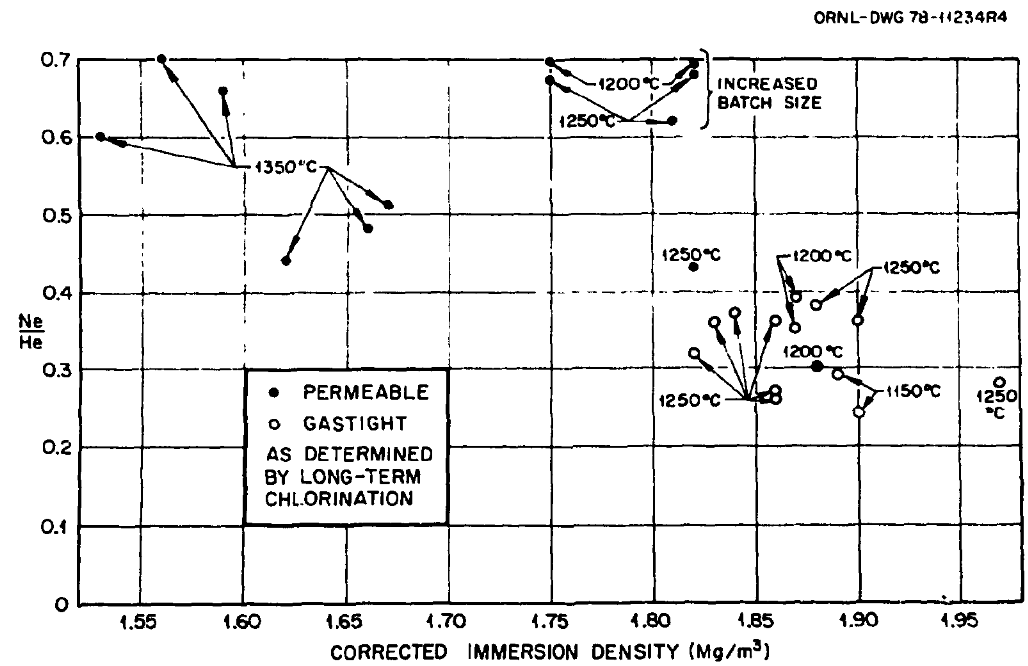

Fig. 7. High Temperatures and Large Batch Sizes Cause Coatings to Be Permeable. Coatings shown here were deposited with a mixture of acetylene and propylene.

permeability. This need increases significantly with coater size. An improved gas distributor was developed at ORNL that produces very uniform fluidization in both 0.13- and 0.24-m coaters.[15]

A final process variable that greatly affects permeability is the coating rate. At low deposition rates coatings tend to be more gastight, as shown in Fig. 8(a). Coating rates must be below 3 μm/min to produce a gastight coating. Coatings deposited at rates between 3 and 4 μm/min must be examined very closely because, depending on the density and porosity, they may be gastight or permeable. Coating rates higher than 4 μm/min usually produce permeable coatings. It is important to remember that the coating rate and temperature are not independent. If the deposition temperature is increased, the coating rate will also increase.

The deposition rate is also an important variable in controlling the anisotropy of the coating. As coating rates increase, individual crystallites deposit in a more random fashion, making more isotropic coatings [Fig. 8(b)]. Normally coating rates above 2.5 μm/min are required to produce an acceptable coating.

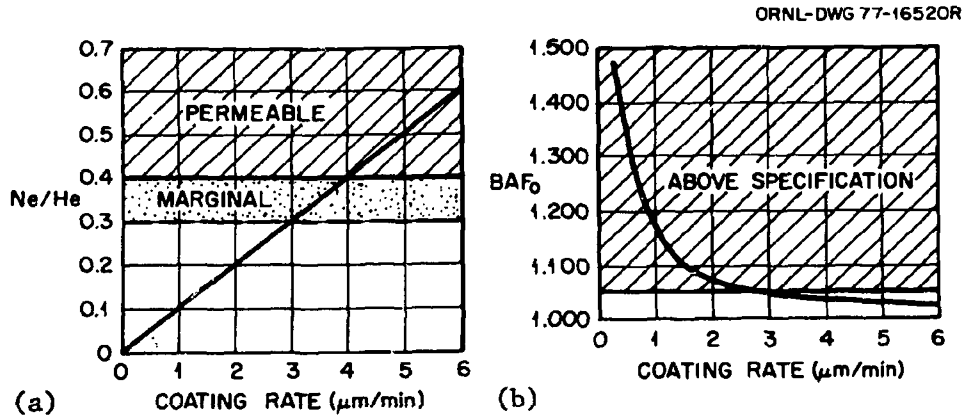

Fig. 8. The Effect of Coating Rates on Permeability. (a) Coatings become permeable as the coating rate increases. (b) However, low coating rates are unacceptable because highly anisotropic coatings result.

At this point it is clear why optimium coatings are so difficult to obtain. The restrictions on coating rate because of anisotropy and permeability reduce the acceptable range of coating rates to between 2.5 and 4 μm/min. Furthermore, the coatings deposited at rates between 3 and 4 μm/min must be carefully examined to assure that they are gastight.

The three techniques used to determine permeability in this work were carefully compared. For Biso-coated particles the neon-helium intrusion and the long-term chlorination techniques correlated excellently. Pyrocarbon coatings with neon-to-helium ratios less than 0.3 (gastight) are also unaffected by the 18-h chlorination (Fig. 9) and are therefore gastight. Coatings with neon-to-helium ratios greater than 0.4 (permeable) show significant migration of heavy metal from the kernel into the buffer layer and are therefore permeable (Fig. 9). Coatings with neon-to-helium ratios between 0.3 and 0.4 can be either gastight or permeable and must be examined by the chlorine leach procedure for the final classification. The fission gas release technique agrees well with the neon-helium and chlorination techniques when the coatings are very permeable. As Fig. 10 shows, fission gas release does not correlate well with the other techniques for slightly permeable coatings. The technique is apparently not sensitive enough to distinguish slightly permeable coatings from gastight ones.

Since both the neon-helium and the chlorine leach techniques determine as-produced permeability, it may be questionable how significant

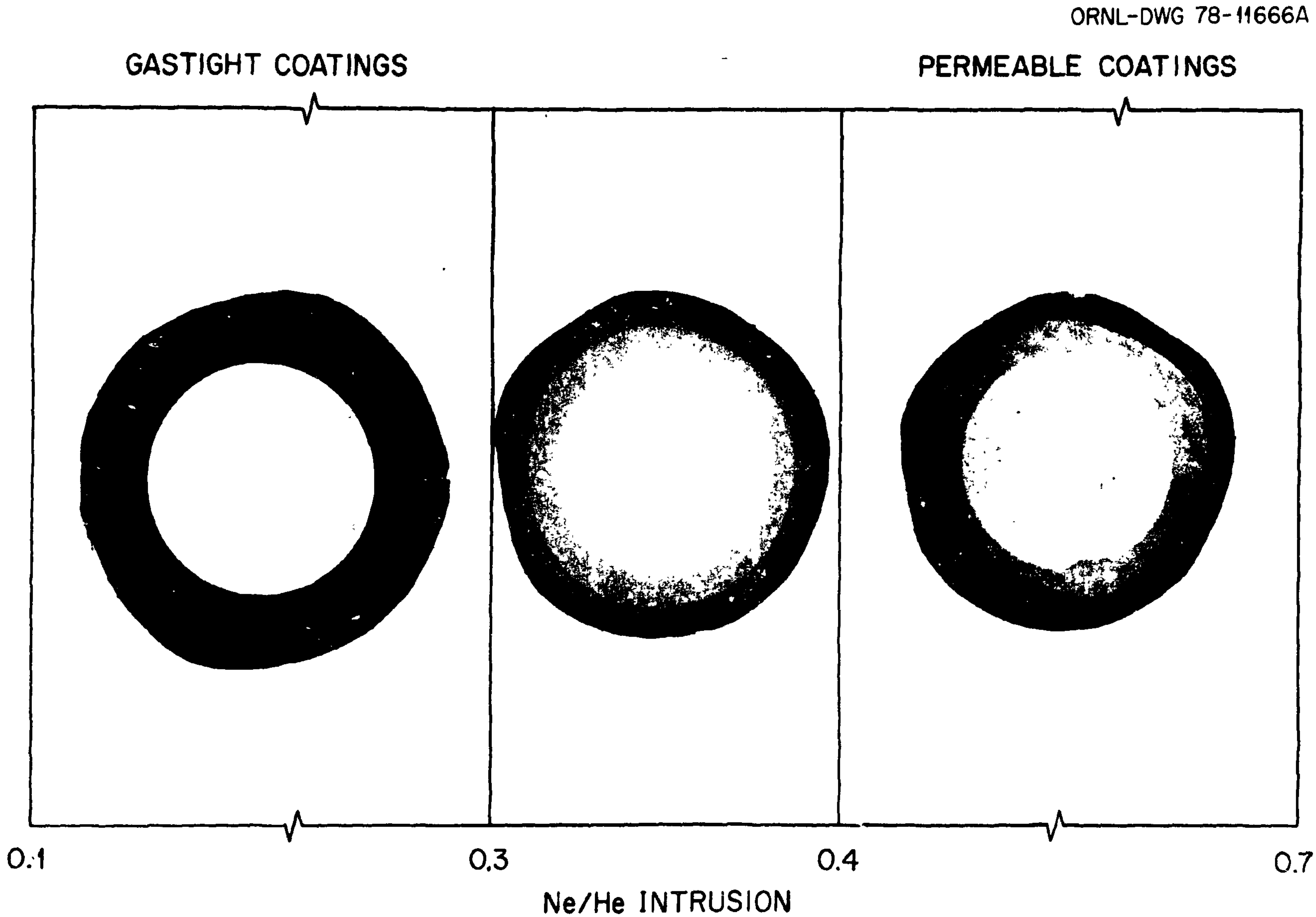

Fig. 9. Particles with Permeable Coatings Can Be Detected by Both Neon-Helium Intrusion and X-Ray Microradiography After Long-Term Chlorine Leach.

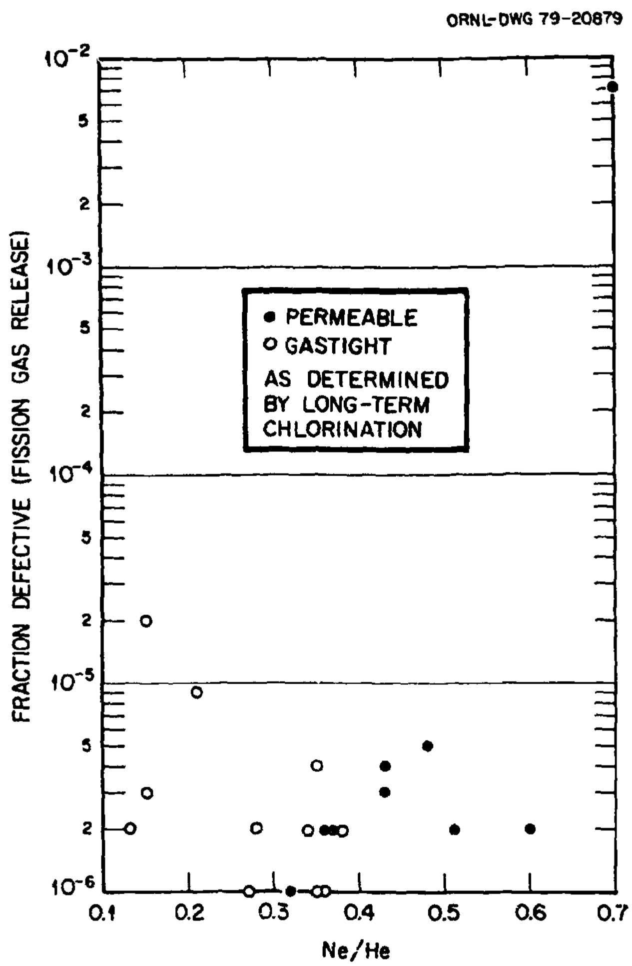

Fig. 10. No Strong Correlation Exists Between Coating Permeability (Ne/He) and the Fraction Defective as Measured by Fission Gas Release.

these results are to the krypton and xenon diffusion through coatings. The fission gases krypton and xenon have atomic radii of 0.19 and 0.21 nm (1.9 and 2.1 Å), respectively. The techniques that have been developed determine if the coatings are permeable to chlorine or to neon. The atomic radii of these two gases are included in the following list:

Gas	Atomic Radii (nm)	(Å)
He	0.14	1.4
Ne	0.16	1.6
Cl	0.18	1.8
Kr	0.19	1.9
Xe	0.21	2.1
$ThCl_4$	0.36	3.6

Since the sizes of these atoms are so close to the size of krypton and xenon, a coating that is permeable to neon or chlorine would probably be permeable to krypton and xenon. However, these results must be correlated with actual fission gas release of krypton and xenon. Valuable information was obtained by comparing preirradiation permeability characterization with postirradiation internal gas inventory measurements and irradiated microsphere gamma analyzer (IMGA) measurements.[16] Internal gas inventory measurements of Biso particles irradiated in irradiation test OF-2 showed that batches with neon-to-helium ratios below 0.3 contained the anticipated amount of fission gases after irradiation. Results obtained by using IMGA analyses showed that a batch with a neon-to-helium ratio of 0.28 also retained the calculated amounts of fission gases. High fission gas release was observed for two driver particle batches used in experiments HRB-7, -8, -9, and -10 (refs. 4 and 17). The Biso particles had neon-to-helium ratios of 0.48 and 0.53. Because of these results the long-term chlorination and neon-helium intrusion are believed to correlate well with in-reactor fission gas release. Biso-coated fuel particle batches with neon-to-helium ratios less than 0.3 show no heavy metal migration after an 18-h chlorination and are gastight to the fission gases krypton and xenon.

CONCLUSIONS

Of the three methods of permeability measurement, the neon-helium technique and long-term chlorination with radiography were most reliable. These methods could easily distinguish permeable coatings from gastight ones. However, if the neon-helium technique yielded values between 0.3 and 0.4, the coating could be permeable or gastight. In this case only the long-term chlorination could distinguish gastight coatings from slightly permeable ones. These techniques have proven very useful in predicting the irradiation behavior of several batches. Additional irradiation tests are being performed to verify the ability of these techniques to predict irradiation behavior.

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