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FISSION-PRODUCT RETENTION IN HTGR FUELS*

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ABSTRACT

Retention data for gaseous and metallic fission products are presented for both Triso-coated and Biso-coated HTGR fuel particles. Performance trends are established that relate fission product retention to operating parameters, such as temperature, burnup, and neutron exposure. It is concluded that Biso-coated particles are not adequately retentive of fission gas or metallic cesium, and Triso-coated particles which retain cesium still lose silver. Design implications related to these performance trends are identified and discussed.

INTRODUCTION

Irradiation testing of nuclear fuels is undertaken to establish performance trends under a variety of operating conditions. Data collected during examination of irradiated fuels form the basis of the fuel performance models which are used to guide core design in a number of areas:

1. selection of operating parameters (burnup, temperature, and flux distributions, temperature gradients, etc.),

2. development of component designs and maintenance philosophy for the primary circuit, consistent with personnel exposure restrictions,

3. development of fuel manufacturing specifications which balance the costs and benefits of improved fuel quality, and

4. calculation of site boundary doses in an operating plant for inclusion in the licensing documents.

The performance attribute of primary importance to the HTGR fuel concept is the ability of the coated particles to retain fission products. The

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postirradiation examination techniques used to measure this capability are well documented[1] and will not be discussed here. Rather, specific measurements of metallic and gaseous fission product inventories from batches of individual coated particles will be presented and the implications for fuel design and reactor operation discussed.

GASEOUS FISSION PRODUCT RETENTION

Measurements indicate that gaseous fission products (krypton and xenon) are generally not sufficiently retained by the pyrocarbon layer in Biso-coated particles. Inventories of fission gas released from kernels have been measured on irradiated coated particles by crushing the particles in an evacuated chamber and measuring the quantities of gas released using a time-of-flight mass spectrometer.[2] Typical data from such measurements are shown in Figs. 1 and 2. In both figures the ^{86}Kr inventories released from the kernels are plotted as a function of position in the irradiation capsule.[3,4] The measured inventories (data points) can be compared with the predicted inventories (cross-hatched areas). The prediced inventories are calculated as discussed in Ref. 5. It is assumed the gas released from a particle, when it is crushed in the postirradiation gas analyzer (PGA) system, is that contained in the buffer layer of the particle. The amount of gas released from the kernel to the buffer during irradiation is a function of the burnup, time, and operating temperature (see Ref. 5). If there is significantly less gas than predicted in the particle when crushed, it is assumed that some gas "leaked out" during irradiation through permeable coatings. This is the case with most of the data presented in Figs. 1 and 2. Most of these data are for Biso-coated particles, where permeable pyrocarbon coating is a problem of major concern.[6]

Several techniques have been developed to measure the degree of pyrocarbon permeability to fission gases prior to irradiation. These techniques attempt to determine the coating properties that influence gas retention in the particle during irradiation. The data in Figs. 1 and 2 show that particles with pyrocarbon coatings characterized as impermeable (solid symbols) performed no better than particles classified as having permeable coatings (open symbols). These results indicate that no reliable characterization technique exists to accurately predict gas retention in Biso-coated particles during irradiation.

The rather poor agreement between measured and calculated gas inventories for the Triso-coated particles in positions 47 and 50 in Fig. 1 deserves mention. We believe the Triso-coated particles are gas retentive. The agreement between measurement and calculation in the high temperature section of that capsule (positions 31 and 37 in Fig. 1) is good. For positions 47 and 50 there is very little scatter in the gas inventory measurements. We believe the calculational model to be deficient under the conditions at the ends of the capsule. This model attempts to calculate gas release from the kernel to the buffer layer as functions of several parameters (burnup, temperature, and irradiation time). At the ends of the capsule temperatures are very uncertain because of axial heat losses and wide swings in neutron flux in the High Flux Isotope Reactor (HFIR). "Average" temperatures here have much less meaning, especially as relating to gas release, than in the

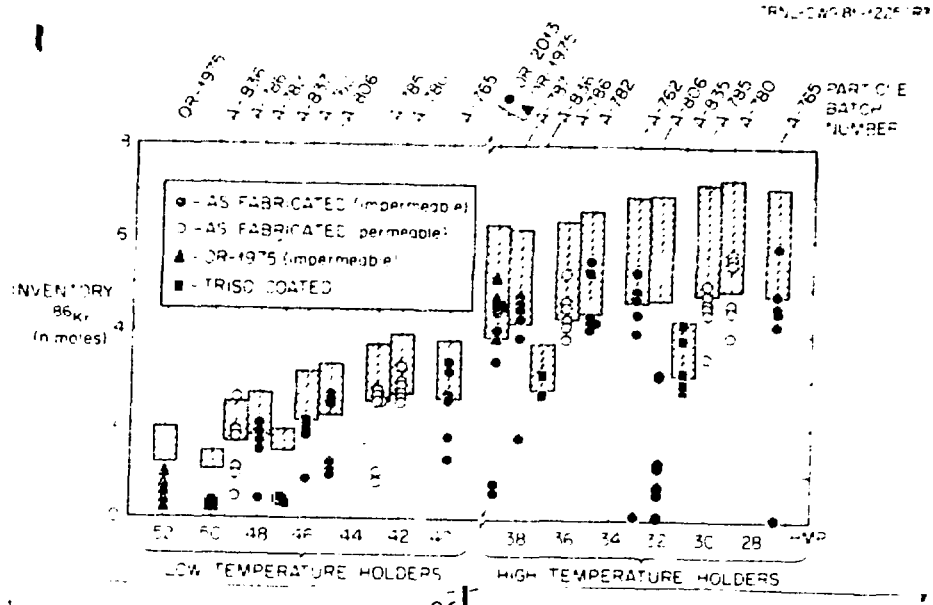

Fig. 1. Comparison of ^{86}Kr fission gas inventory measured with PGA system with predicted inventory released from kernel to buffer (shaded area). Data from HT-34 experiment (Ref. 3). Peak particle temperatures in low-temperature magazine were about 1160°C; peak temperatures in high-temperature magazine were about 1410°C.

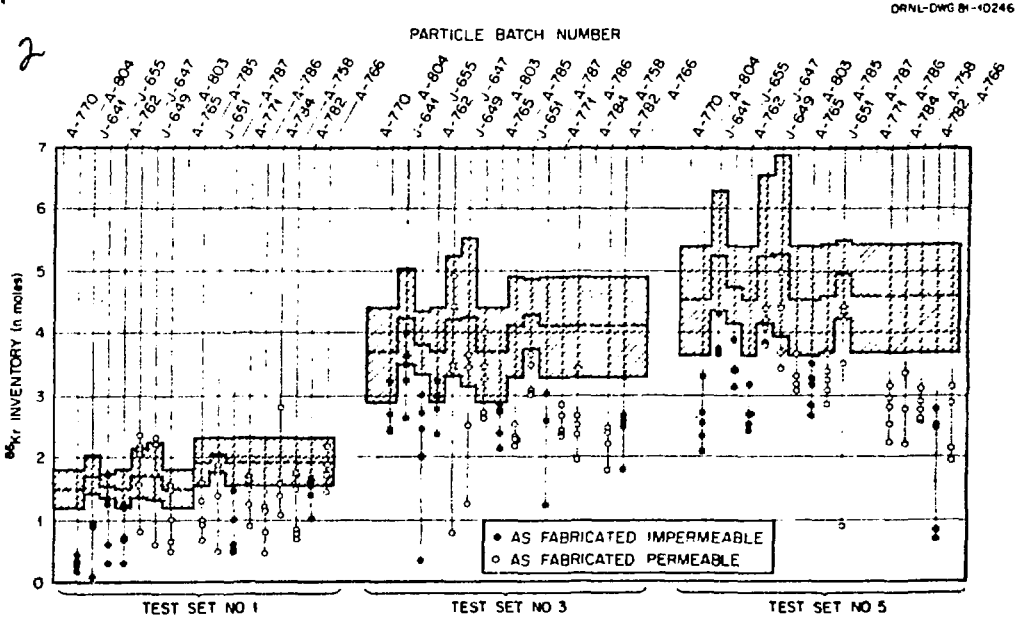

Fig. 2. Comparison of ^{86}Kr fission gas inventory measured with predicted inventory released from kernel to buffer (shaded area). Data from HRB-14 experiment (Ref. 4). Peak design temperature for this capsule was 1250°C.

center of the capsule. We believe that the disagreement between measurement and calculation is due to an overprediction (by the calculation) of the amount of gas released from the kernel to the buffer.

The large amount of "within-batch" scatter in the gas retentiveness of the pyrocarbon coatings shown in Figs. 1 and 2 was unexpected. The "within-batch" scatter is larger than the "between-batch" scatter and makes batch-to-batch comparisons difficult. It has been postulated that performance variation is due to property variation, which occurs as a result of the random manner in which coatings are deposited in a fluidized bed. Coating rate has been shown to be a very important parameter[7] in influencing coating properties. Coating rates are determined by measuring an "average" coating thickness (from many particles) and dividing by the duration of the coating run. If one hypothesizes two particles with the same coating thickness, where one particle alternated between positions of maximum coating gas concentration (rapid coating rate) and positions at the edge of the coater (very low coating rate), and the other particle occupied positions of intermediate coating rate during the run, it is easily concluded that the two particles would have coatings with very different properties, but the coating rates would be calculated to be the same. It could now be expected that the performance of the two coatings during irradiation might differ significantly.

An indication of the range of property variation within as-coated batches of Biso-coated particles is shown in Fig. 3. Preirradiation property characterization data are presented for the batches for which gas content measurements are shown in Figs. 1 and 2. Significant variations in the Ne/He ratios are evident. The Ne/He measurement was developed to be an indication of coating permeability. The measurement is made by comparing the permeability of the coating to the relatively large Ne atom with the small He atom.[7] The permeable/impermeable classifications shown in Figs. 1 and 2 were made on the basis of the "average" values shown in Fig. 3. However, the range of values shown in Fig. 3 illustrate that some particles from each batch extend well into both classifications. F3

The BAF_0 measurement is an indication of the amount of crystallite disorder in the coating. Highly ordered (high BAF_0) crystallite structures are gastight in the as-fabricated condition but develop microcracks during irradiation, and thus lose fission gas. Highly disordered (low BAF_0) structures withstand irradiation damage better than the highly ordered structures, but are permeable to fission gas. An intermediate degree of disorder was considered desirable to be resistant to irradiation damage and remain gastight during irradiation.[5] The "window" of intermediate BAF_0 has been shown to be very narrow.[7] The within-batch scatter in BAF_0 values shown in Fig. 3 exceeds the width of this window for most batches.

METALLIC FISSION PRODUCT RETENTION

Measurement of metallic fission product inventories in coated particle fuels has focused mainly on cesium and silver. Cesium is the most abundant volatile metallic fission product, and silver is an extremely volatile activation product produced in much smaller quantities. Both cesium and silver

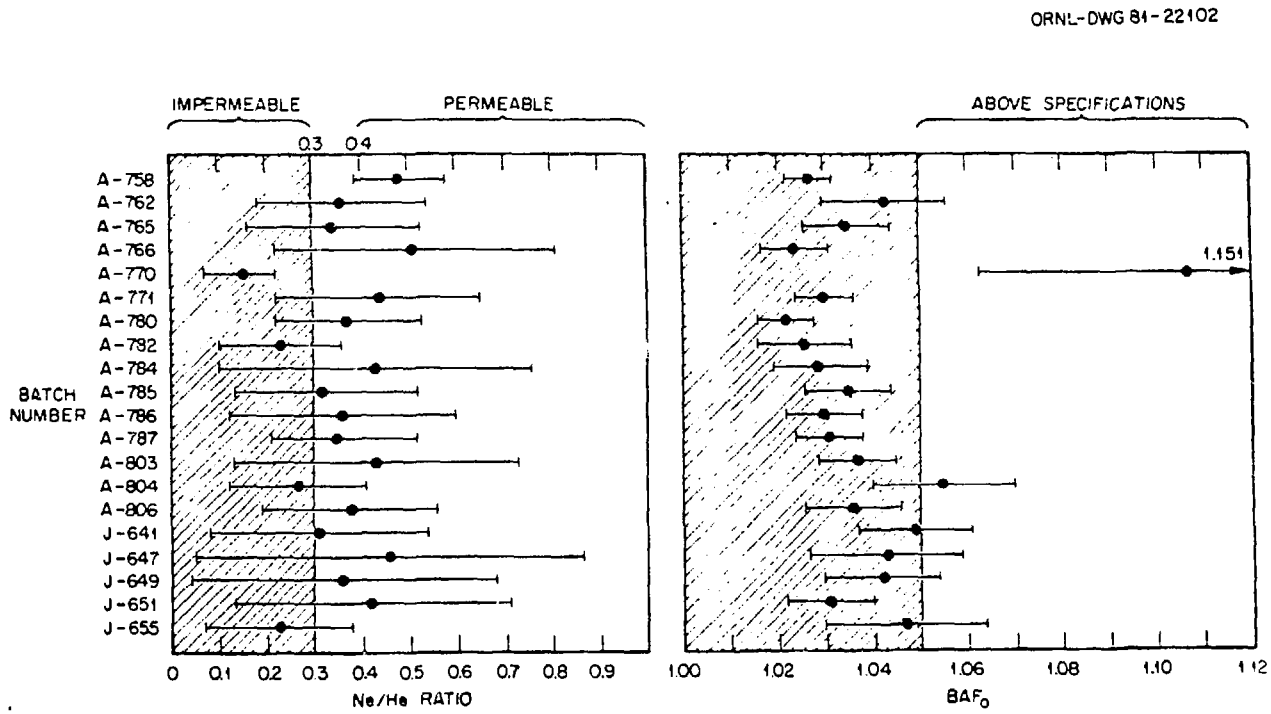

Fig. 3. Summary of coating characterization data for Biso-coated fertile particle batches irradiated in capsules HT-34 and HRB-14. Data show within-batch property variations are greater than batch-to-batch variation.

plate out on cooler surfaces within the primary circuit and introduce significant maintenance problems.

Metallic fission product inventories are measured using the irradiated microsphere gamma analyzer (IMGA) system.[8-10] Relative inventories of volatile fission and activation products are based on gamma activity ratios between volatile (^{137}Cs, ^{110m}Ag) and nonvolatile (^{95}Zr, ^{106}Ru) species. Using ratios compensates for the slight variation in kernel diameter (fission product inventory) between particles. Typical data from IMGA in histogram form are shown in Fig. 4 for Triso-coated UC_2 with a ZrC layer used in place of the SiC layer in the standard Triso particle design. These particles were irradiated in capsule HRB-15a at "normal" HTGR operating temperatures (1150 to 1250°C). Shown there are three histograms. Care must be taken in comparing them because of different ranges in the X-axes. The X-axis is 50 channels wide in each case and covers the full range of activity ratios measured for the total population. The particle batch examined contained 342 Triso-coated particles. Channel 1 in each histogram is designated by the lowest ratio measured, and channel 50 is designated by the highest ratio. The channel width is determined by the range between the highest and lowest ratio. The Y-axes represent the number of particles per channel. The histogram of $^{95}Zr/^{106}Ru$ (left of Fig. 4) is a very tight distribution compared to the others. The range in this measurement is from 97 to 108% of the mean. This histogram of the ratio of two nonvolatile fission products gives a rough idea of the burnup distribution due to the flux gradient in which the fuel was irradiated. The activity ratios are strongly burnup dependent. F4

The other two histograms represent the ratio of a volatile fission product to a nonvolatile fission product. The difference in the shape of the two histograms is significant. The mean of the center distribution represents only 55% of the calculated ^{110m}Ag inventory (calculated inventory is based on neutronics data coupled with depletion computer codes). The range extends from 0 to 73%, which indicates that even the best performing particles have lost about 25% of their ^{110m}Ag inventory. The $^{137}Cs/^{106}Ru$ histogram contains a "normal" distribution to the right with several "blips" scattered to the left. These "blips" (perhaps 15 in all) are classified as "failed" particles because they have lost a significant fraction (up to 93%) of their ^{137}Cs inventory. The mean of the $^{137}Cs/^{106}Ru$ distribution represents 99% of the calculated ^{137}Cs inventory. If the failed particles were not included the mean would be about 100%. It is postulated that ^{137}Cs escaped through cracked or otherwise damaged coatings. Comparing the $^{110m}Ag/^{106}Ru$ histogram with the $^{137}Cs/^{106}Ru$ histogram, it is evident that ^{110m}Ag escaped from particles that contained all ^{137}Cs. Silver apparently diffuses through intact coatings.

The trends shown in Fig. 4 have also been observed with the "standard" fissile particle design (SiC interlayer). Figure 5 shows data taken from planchet 81 irradiated in capsule HRB-14. Of the approximately 30 Triso-coated fissile particles examined, all retained essentially 100% of the ^{137}Cs, while ^{110m}Ag retention ranged from 0 to about 75%. F5

Capsules HRB-14 and -15a were operated at "normal" HTGR temperatures, and ^{110m}Ag loss was significant. Another capsule (HRB-15b) was operated at

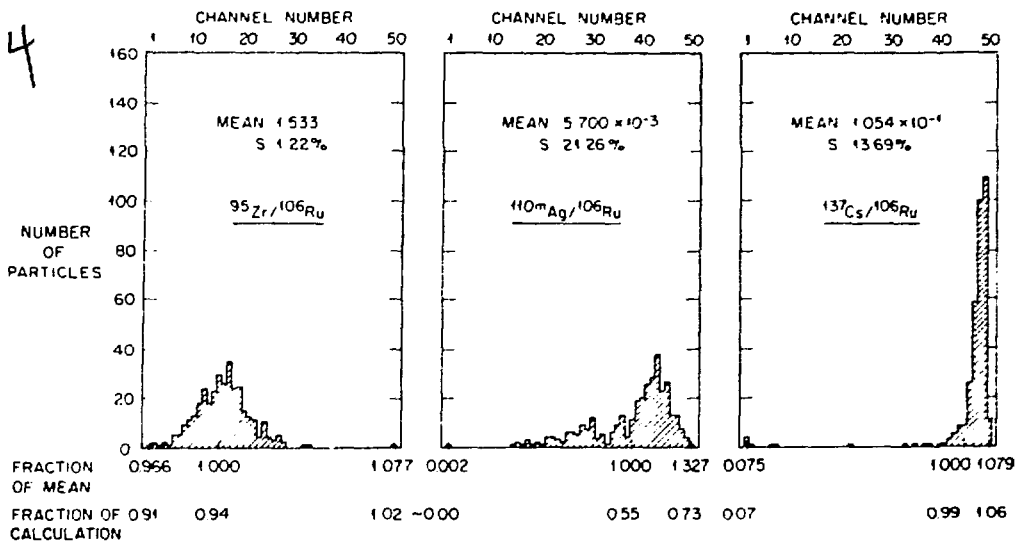

Fig. 4. $^{95}Zr/^{106}Ru$, $^{110m}Ag/^{106}Ru$, and $^{137}Cs/^{106}Ru$ histograms for 342 irradiated particles examined with the IMGA system. Triso-coated UC_2 (with ZrC replacing the SiC) from batch 6161-00-010 irradiated in capsule HRB-15a (rod 7) at an average temperature of 1191°C.

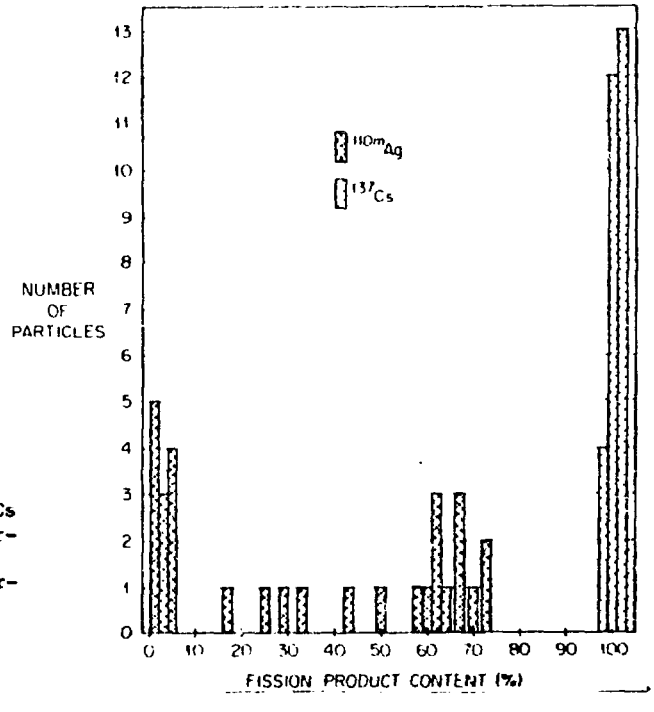

Fig. 5. Histograms of ^{110m}Ag and ^{137}Cs retention data for Triso-coated fissile particles irradiated in planchet 81, capsule HRB-14. The design peak irradiation temperature was 1250°C. Data show Triso-coated particles which retained 100% cesium lost significant silver.

temperatures significantly below normal (in the range of 900 to 1000°C), and ^{110m}Ag loss was also significant.[11] Even at these low temperatures ^{110m}Ag retention ranged from 68 to 78%.

Attempts to describe ^{110m}Ag retention in standard Triso-coated particles as functions of temperature and neutron exposure are described in Figs. 6 and 7. Figure 6 shows data from three irradiation capsules with operating temperatures ranging from about 900 to 1400°C. Some of the scatter in this plot is undoubtedly due to the broad range of temperatures. Retention of ^{110m}Ag appears to diminish significantly with increasing fast-neutron fluence over this temperature range. In Fig. 7 the results of a regression analysis are shown, where the influence of temperature and fluence on the ^{110m}Ag diffusion coefficient in SiC is described. These limited data suggest that the influence of neutron fluence diminishes with increasing temperature, as the diffusion coefficient increases rapidly with increasing temperature above about 1100°C.

Metallic fission product retention in Biso-coated fertile particles has also been investigated. Little ^{110m}Ag data have been collected for thorium fuels because ^{110m}Ag inventories in such particles are extremely low. This is because the fission yields of ^{109}Ag are 25 to 45 times lower for ^{233}U fissions in fertile fuel than for the ^{239}Pu and ^{241}Pu fissions in the 20% enriched fissile fuel [^{110m}Ag is an activation product of the stable isotope ^{109}Ag (^{109}Ag [n,γ]^{110m}Ag)] and the burnups are about one-third the fissile particle level in the fertile fuel. Retention of ^{137}Cs has been studied extensively. Figures 8 and 9 show the results of ^{137}Cs retention measurements on Biso-coated fertile particles irradiated in capsules HT-34 and HRB-14. These are the same particles for which fission gas retention measurements were presented in Figs. 1 and 2. As shown, ^{137}Cs retention is good for particles irradiated in the low temperature section of capsule HT-34. However, particles irradiated in the high temperature section of HT-34 and in capsule HRB-14 lost large fractions of their ^{137}Cs inventories. Particles irradiated in the high temperature section of capsule HT-34 (at average temperatures 100 to 200°C higher than the particles irradiated in capsule HRB-14) lost 49 to 90% of their ^{137}Cs, while the HRB-14 particles lost 0 to 60%. There is considerable scatter in the measurements in both data sets. This scatter is also attributed to the within-batch scatter in coating properties, discussed earlier.

DISCUSSION AND CONCLUSIONS

A significant body of very pertinent data is being developed for the HTGR core designers on gaseous and metallic fission product retention of coated particle fuels. Some important conclusions have been drawn from this work and parallel programs in other countries (the United Kingdom, Federal Republic of Germany, France, and Japan).

- Biso coatings cannot contain sufficient cesium at operating temperatures of interest for the U.S. HTGR design.

- Triso coatings can contain cesium as long as they remain intact.

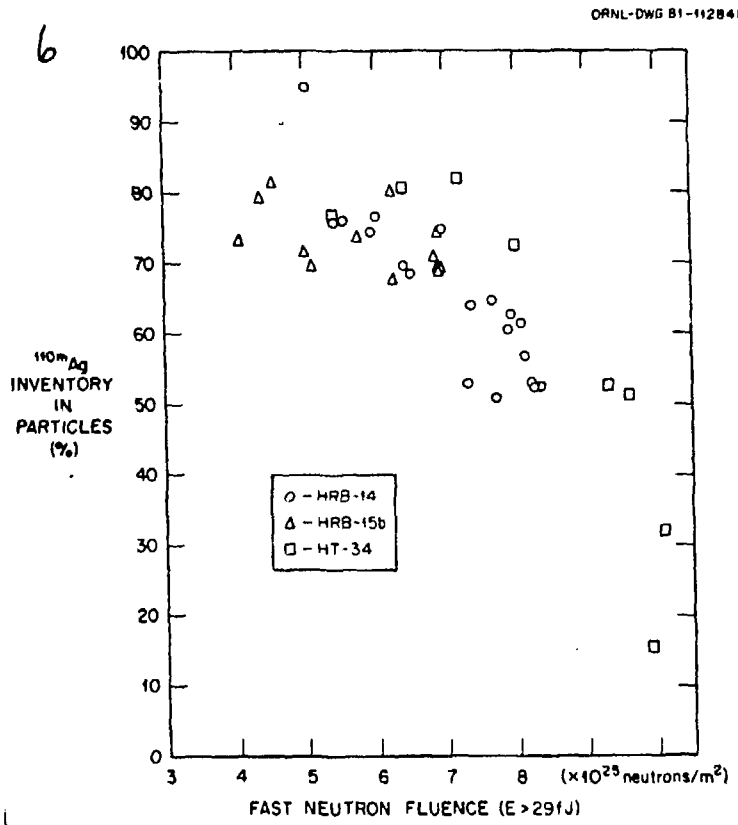

Fig. 6. ^{110m}Ag retention as a function of fast neutron fluence for particles irradiated in several irradiation capsules. Data show ^{110m}Ag retention decreases with increasing fluence. Scatter in the data is due in part to differences in operating temperature and burnup.

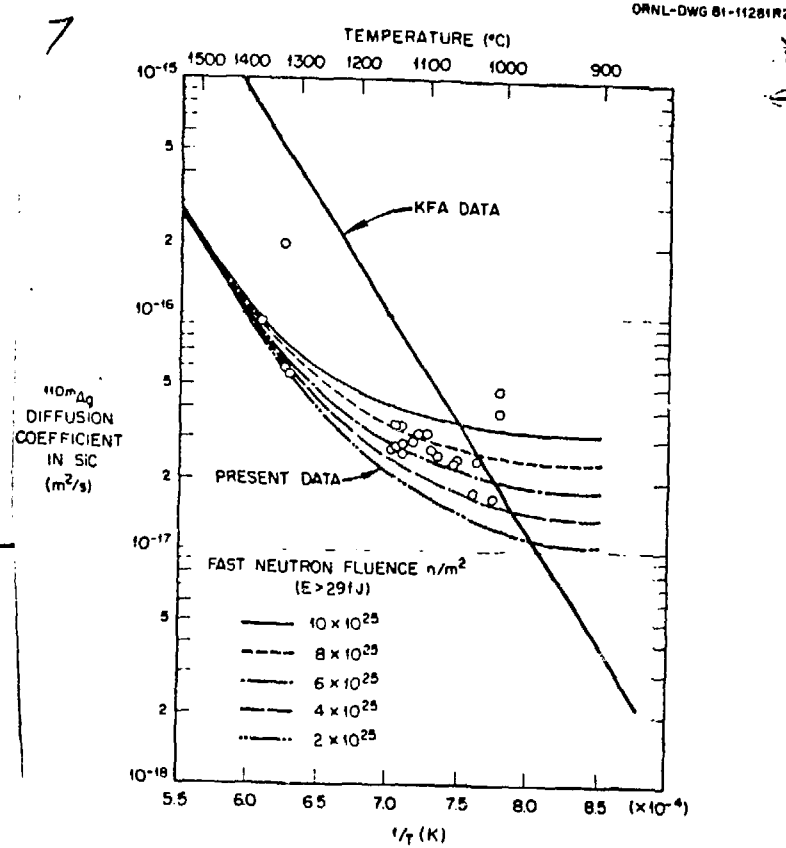

Fig. 7. ^{110m}Ag diffusion coefficient in SiC as a function of reciprocal temperature for particles irradiated in several capsules. Data show strong neutron dependence at lower temperature.

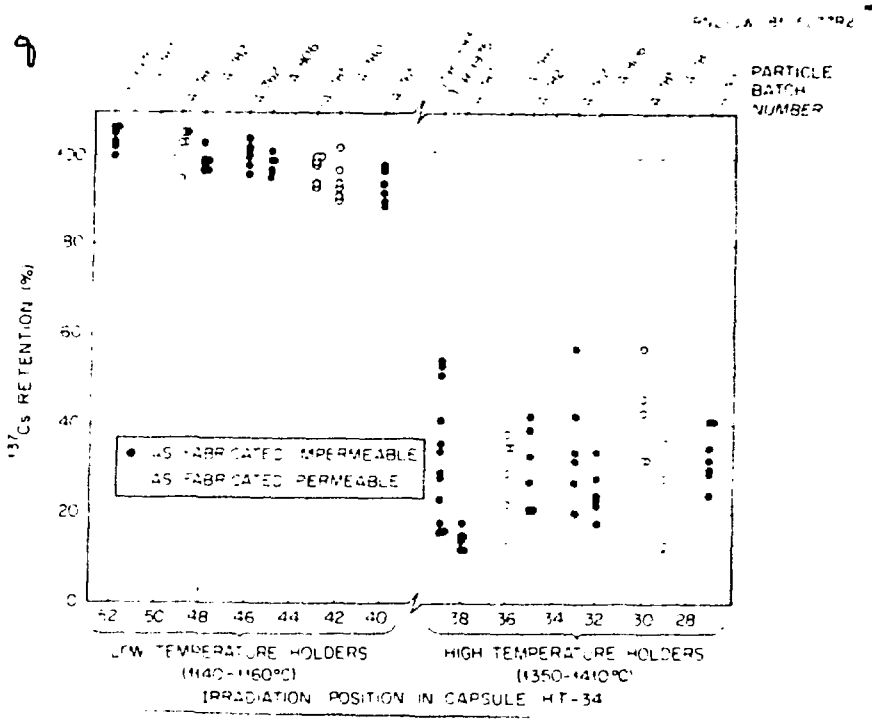

Fig. 8. ^{137}Cs retention data for Biso-coated fertile particles irradiated in capsule HT-34. Data show good retention in low temperature section of capsule and poor retention in high temperature section.

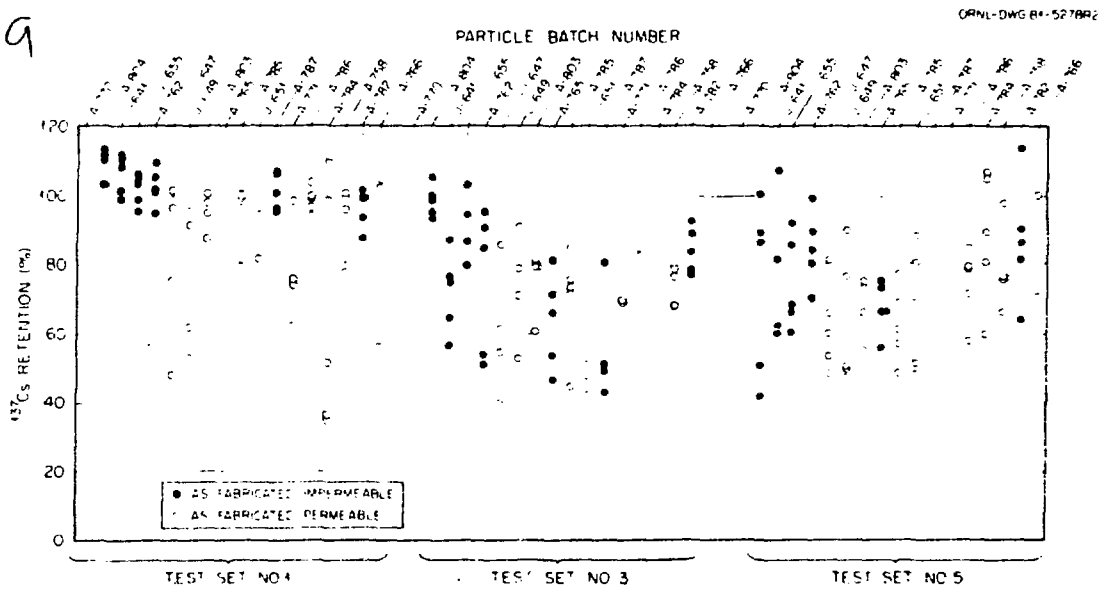

Fig. 9. ^{137}Cs retention data for Biso-coated fertile particles irradiated in capsule HRB-14. Data show considerable scatter in retentiveness in all but the top end of the capsule.

- Biso coatings can be fabricated that are impermeable to fission gas during irradiation, but such coatings cannot be produced economically in large batches with current technology.

- Techniques do not exist for characterization of Biso coatings so as to reliably predict fission gas retention during irradiation.

- SiC coatings cannot contain all of the ^{110m}Ag produced during irradiation, even at irradiation temperatures as low as 900°C. Silver release increases with increasing temperature and neutron fluence.

Design changes have been made to respond to what has been learned about fission product retention in coated particle fuels:

- The Biso particle design for fertile fuel has been abandoned in favor of the more retentive Triso design.

- Design changes have been made in the fuel element to reduce operating temperatures (smaller fuel rods and more fuel holes).

- The maximum neutron fluence has been reduced from about 8×10^{25} neutrons/m^2 ($E > 29$ fJ) for the highly enriched uranium (HEU) fuel cycle to about 6.5×10^{25} for the low enriched uranium (LEU) fuel cycle.

- The fuel performance models indicate that silver release is not a major problem for the steam cycle/cogeneration HTGR, where only a small fraction of predicted worker exposure comes from metallic fission products plated out in the primary circuit. Only a fraction of the predicted plateout activity comes from silver. However, for higher technology designs, the predicted plateout exposures are more severe, and remotely maintained systems will be considered.

- Core outlet temperatures (COT) are being reduced so as to minimize peak fuel temperatures. The current design COT for the steam cycle/cogeneration HTGR is about 680°C compared to the COT 750°C for the Fort St. Vrain reactor.

Assuming the design changes detailed above, the core performance models predict the peak fuel temperatures are reduced from about 1350°C to about 1250°C, with a corresponding reduction of approximately 100°C in the average fuel temperature. Calculated core performance under these conditions is such that the maximum permissible site boundary doses are not exceeded, and the primary circuit remains clean enough to permit contact maintenance. All of the above comments apply to the 2240 MW(t) steam cycle/cogeneration HTGR. For the 1170 MW(t) process heat HTGR (COT 950°C) the operating temperature calculated is somewhat higher (about 1315°C peak), but the fuel residence time is three years instead of four.

Continued irradiation testing of the reference fuel through the qualification and licensing phase will provide additional fission product release data to improve and validate the fuel performance models.

REFERENCES

1. F. J. HOMAN, M. J. KANIA, and O. M. STANSFIELD, "Postirradiation Characterization and Performance Assessment of Coated Particle Fuels," Proceedings of British Nuclear Energy Society Conference on *Gas-Cooled Reactors Today*, 20–24 September 1982 (to be published).

2. M. J. KANIA and E. L. LONG, JR., "Gas Content Measurements of Irradiated Coated Particles Using the PGA System," Proceedings of British Nuclear Energy Society Conference on *Post-Irradiation Examination*, 13–16 May 1980, p 255, London (1980).

3. E. L. LONG, JR., et al, *Performance of HTGR Biso- and Triso-Coated Fertile Particles Irradiated in Capsule HT-34*, Oak Ridge National Laboratory ORNL/TM-7674 (August 1981).

4. *High-Temperature Gas-Cooled Reactor Technology Development Program Annual Progress Report for Period Ending December 31, 1980*, Oak Ridge National Laboratory, ORNL-5753 pp 28–31.

5. M. J. KANIA, F. J. HOMAN, and B. A. THIELE, *Irradiation-Induced Permeability in Pyrocarbon Coatings — Final Report of Work Conducted Under PWS FD-12*, Oak Ridge National Laboratory, ORNL/TM-8348 (in press).

6. R. A. BRADLEY and B. A. THIELE, "Neutron-Induced Permeability of Pyrocarbon-Coated High-Temperature Gas-Cooled Reactor Fuel Particles," *Nuclear Technology*, 35(2), pp 353–358 (September 1977).

7. D. P. STINTON, et al, *Detection and Control of As-Produced Pyrocarbon Permeability in Biso-Coated High-Temperature Gas-Cooled Reactor Fuel Particles*, Oak Ridge National Laboratory, ORNL/TM-7254 (May 1980).

8. K. H. VALENTINE and M. J. KANIA, *IMGA Operating Manual*, Oak Ridge National Laboratory, ORNL/TM-6567 (August 1979).

9. M. J. KANIA, T. N. TIEGS, and K. H. VALENTINE, *The Irradiated-Microsphere Gamma Analyzer (IMGA) — An Integrated System for HTGR Coated Particle Fuel Performance Assessment*, Oak Ridge National Laboratory, ORNL-5606 (February 1980).

10. M. J. KANIA, T. N. TIEGS, and K. H. VALENTINE, "Irradiation Performance Assessment of Coated Particle Fuels Using the IMGA System," Proceedings of British Nuclear Energy Society Conference on *Post-Irradiation Examination*, 13–16 May 1980, p 232, London (1980).

11. *High-Temperature Gas-Cooled Reactor Technology Development Program Annual Progress Report for Period Ending December 31, 1980*, Oak Ridge National Laboratory, ORNL-5753 pp 83–90.