Post-Irradiation Examination of ^{237}Np Targets for ^{238}Pu Production

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Abstract[1]. Oak Ridge National Laboratory is recovering the US ^{238}Pu production capability and the first step in the process has been to evaluate the performance of a ^{237}Np target cermet pellet encased in an aluminum clad. The process proceeded in 3 steps; the first step was to irradiate capsules of single pellets composed of NpO_2 and aluminum power to examine their shrinkage and gas release. These pellets were formed by compressing sintered NpO_2 and aluminum powder in a die at high pressure followed by sintering in a vacuum furnace. Three temperatures were chosen for sintering the solution precipitated NpO_2 power used for pellet fabrication. The second step was to irradiate partial targets composed of 8 pellets in a semi-prototypical arrangement at the two best performing sintering temperatures to determine which temperature gave a pellet that performed the best under the actual planned irradiation conditions. The third step was to irradiate ~50 pellets in an actual target configuration at design irradiation conditions to assess pellet shrinkage and gas release, target heat transfer, and dimensional stability. The higher sintering temperature appeared to offer the best performance after one cycle of irradiation by having the least shrinkage, thus keeping the heat transfer gap between the pellets and clad small minimizing the pellet operating temperature. The final result of the testing was a target that can meet the initial production goals, satisfy the reactor safety requirements, and can be fabricated in production quantities. The current focus of the program is to verify that the target can be remotely dissembled, the pellets dissolved, and the ^{238}Pu recovered. Tests are being conducted to examine these concerns and to compare results to code predictions. Once the performance of the full length targets has been quantified, the pellet ^{237}Np loading will be revisited to determine if it can be increased to increase ^{238}Pu production.

Keywords: Post-Irradiation, ^{237}Np target, ^{238}Pu, cermet pellet.

TARGET PELLET AND POST IRRADIATON PATH

A three step process has been chosen for the recovery of the US ^{238}Pu production capability which incorporates a graded approach to provide high confidence to the High Flux Isotope Reactor (HFIR) operator that the probability of developmental target failure is low [1]. The 3 steps are:

1. Irradiation of robust single pellet capsules for initial examination of pellet gas release and pellet dimensional change. These capsules were irradiated for 1 and 2 cycles in the HFIR;
2. Partially loaded targets, containing 8 pellets, in a prototypic target configuration were irradiated for first 1 cycle and then 2 cycles in the HFIR. The targets were punctured for gas release measurements and disassembled for individual pellet dimensional measurements;

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3. Fully loaded prototypic targets, containing approximately 50 pellets, were irradiated for first 1 cycle and then 2 cycles in the HFIR. The targets were punctured for gas release measurements and disassembled for individual pellet dimensional measurements.

The pellets were formed by compressing sintered NpO_2 and aluminum powder in a die at high pressure followed by sintering in a vacuum furnace. Three temperatures were chosen for sintering the solution precipitated NpO_2 power used for pellet fabrication. A goal of the irradiation testing was to examine the effects of NpO_2 sintering temperature on pellet in-pile behavior and chemical process solubility. Pellet pressing pressures were as high as practical consistent with reasonable die and punch life. High NpO_2 sintering temperatures offer less shrinkage during irradiation; this results in smaller pellet/cladding gaps and better heat transfer with the resulting low operating temperatures. Low temperatures generally give less gas releases and greater reactor safety margins. However, these high sintering temperatures also make the irradiated pellets more difficult to dissolve and thus complicate the downstream chemical processing required to recover the plutonium.

After loading the targets with the pellets and welding shut, the completed target was hydrostatically compressed to reduce the pellet/clad gap to the minimum practical to give the pellet fabricators as much design/processing leeway as possible.

SINGLE PELLET CAPSULE POST IRRADIATON EXAMINATION

A total 14 single pellet capsules underwent post-irradiation examination (PIE). The capsule design was very robust to contain the fission gases and any pellet debris should problems have arisen. The primary goal was to measure gas release and pellet dimensional change on the first batch of pellet designs to gain general information and to reduce the field of candidates for the partial length targets. The capsule contained the test pellet in an aluminum mount with an upper copper heat sink spring loaded to provide firm contact surface for heat transfer. The design of the single pellet capsule is shown in Figure 1.

After irradiation, the capsules were transferred to the Irradiated Fuels Examination Laboratory (IFEL) for remote examination and disassembly. After an initial visual examination, the capsule was prepared for gas puncture by trimming the capsule wall thickness down so that it could be punctured by a hardened steel punch. After trimming, the capsule was placed into a puncture unit, the unit evacuated, sealed, and then punctured. The released gas was then routed, by a helium sweep gas, into a liquid nitrogen cooled charcoal cold trap where the fission gases were frozen out. After the system had been sweep out by the flowing He, the cold trap was removed from the flow path and gamma counted to determine the ^{85}Kr, ^{131m}Xe, and ^{133}Xe inventories. The puncture unit was also used to measure the capsule plenum volume; an abnormal volume would have been an indication of unexpected behavior. No unusual measurements were noted. Finally, the capsule was opened so that the pellet and internal components could be removed. This sequence of operations is shown in Figure 2. All work was done remotely because of the very high radiation levels.

Gas release varied over a wide range, but was less than 5% (^{85}Kr). After removal, each pellet was measured for length and diameter and the dimensional measurements indicated that the pellets shrunk by 1-6% in volume. The pellets with the higher NpO_2 sintering temperatures shrunk less overall. A metallographic mount was prepared of one of the pellets and it appeared some of the pellet shrinkage was due to the irradiation induced sintering of the NpO_2 particles. The particles appear to be shrinking and drawing away from the aluminum matrix leaving a small gap. It is likely that a small amount of additional matrix sintering is taking part as well, with the net result that early in the irradiation the pellets are increasing in density and thus a reduction in volume is taking place. Later on, much later in the irradiation, one would expect the pellets to begin swelling as fission products accumulate from parasitic fissions. The NpO_2 powder with the higher sintering temperature is denser to begin with and thus suffers less in-pile sintering and volume reduction. A before irradiation and after irradiation micrograph is shown in Figure 3. An evaluation of the irradiation parameters and the observed pellet performance indicated to the program that the maximum acceptable NpO_2 powder sintering temperature and maximum pellet pressing pressures along with the existing fabrication pellet sintering schedule would provide best target pellet.

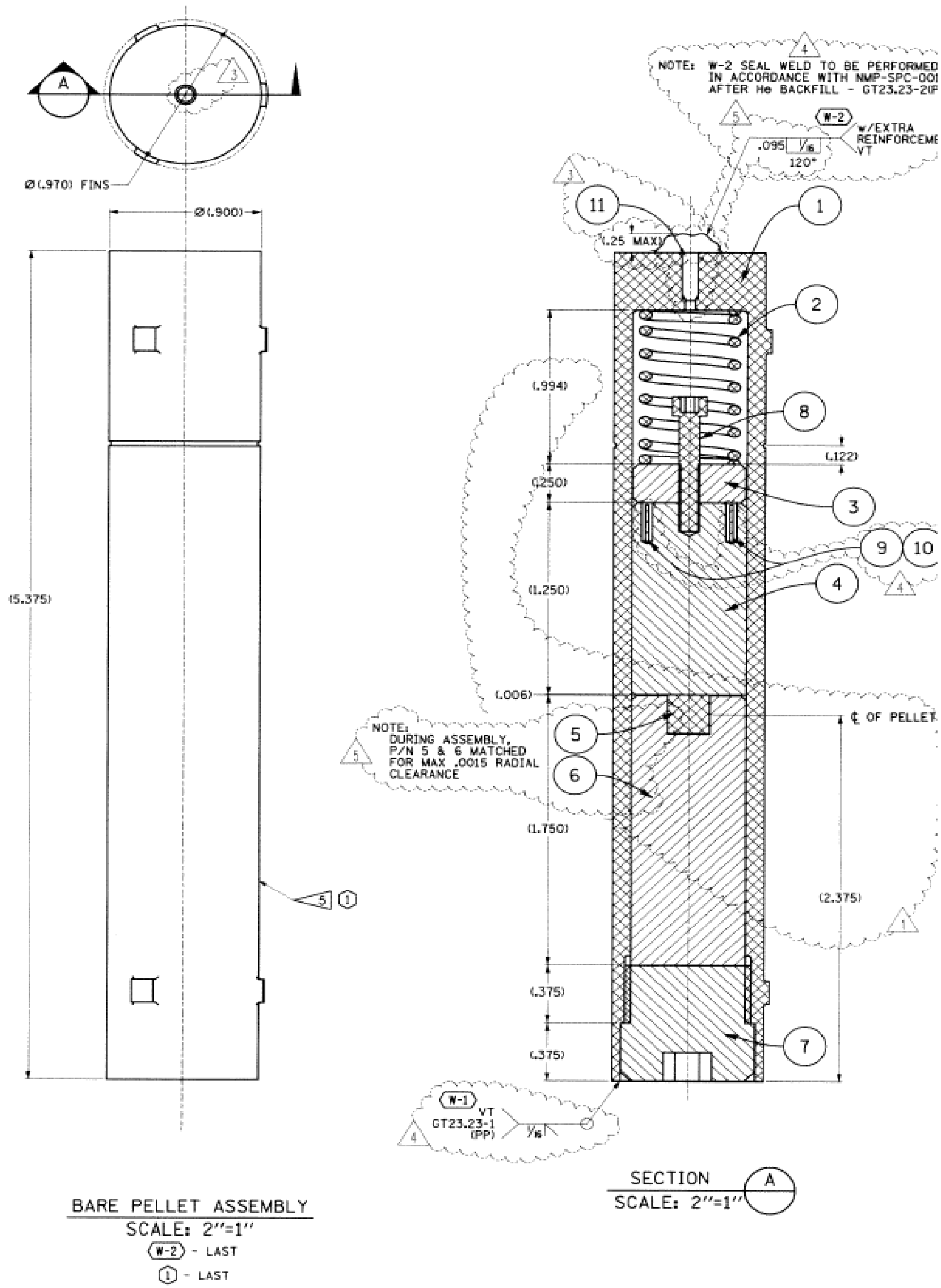

Figure 1. Robust single pellet capsule for initial testing. Item 5 is the pellet, 6 is the aluminum mount, and 4 is the copper heat sink.

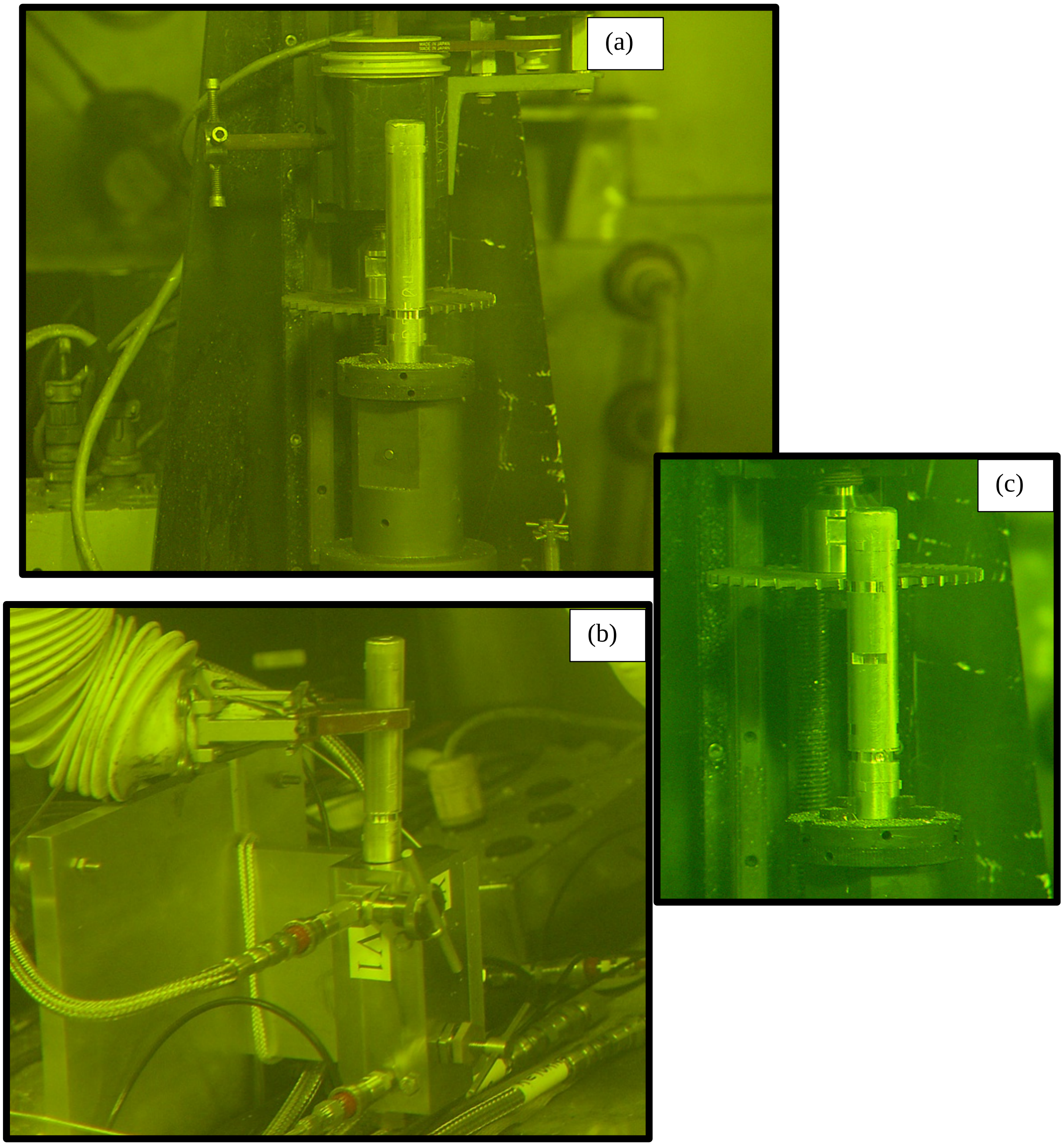

Figure 2. Initial capsule handling: (a) trimming the capsule wall for gas puncture; (b) placing the capsule in the gas puncture unit; (c) cutting the capsule open.

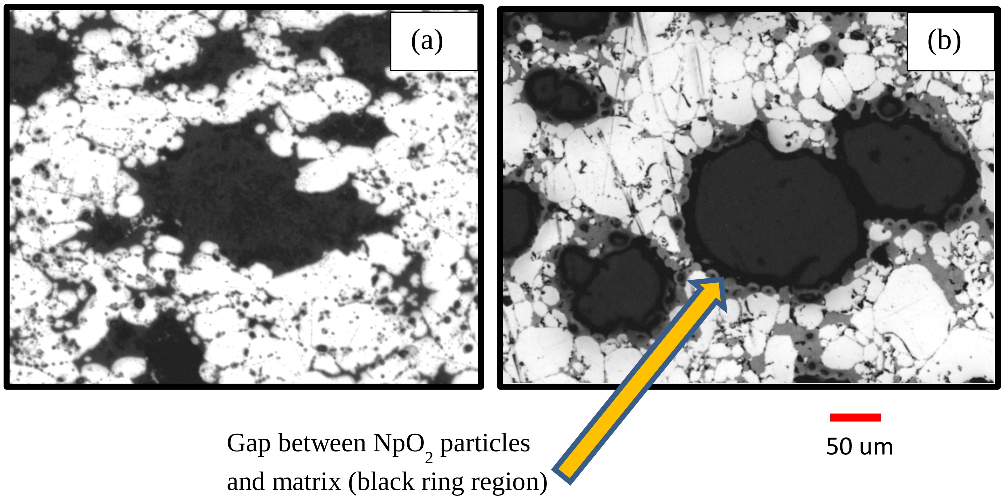

Figure 3. Metallographic mount of pellet showing small gap between the aluminum matrix and the NpO_2 particles due to in-pile particle sintering.

In addition to the pellet selection, the cladding performance for the candidate target design was also examined. Three capsules of similar design were constructed with sets of aluminum alloy tensile test coupons, some in direct contact with special wafer pellets, for irradiation testing of the cladding. These capsules were irradiated in a similar fashion as the single pellet capsules; see Figure 4.

After irradiation, the capsules were opened and the tensile specimens removed and visually examined; nothing unusual was noted. The specimens were then tensile tested and their tensile strength was found to have increased and their ductility decreased in accordance with expectations, both for the specimens in contact with the pellets and those not in contact with the pellets. The material irradiation changes were consistent with the literature and the cladding performance met the needs of the target irradiation; no further cladding test were needed or conducted.

PARTIAL LENGTH TARGET POST IRRADIATON EXAMINATION

A total 10 partial length targets underwent PIE. These were nearly prototypical targets containing 8 pellets in the central region of the target with spacers to fill the remaining volume in the target. Two ^{237}Np sintering temperatures were tested, the lowest having been eliminated by the single capsule testing. The central region of the target has a nearly homogeneous radiation environment so the 8 pellets saw the same irradiation conditions and thus functioned as 8 identical specimens for data collection. Both 1 and 2 irradiation cycle partial length targets were examined in the hot cell.

These targets underwent gas puncture and analysis in a manner that was similar to that of the single pellet capsules; the ^{85}Kr gas release fraction varied over a wide range, 1-13% due to different ^{237}Np sintering temperatures and irradiation conditions depending on the target location in the reactor. After disassembly, the pellets were measured and found to have shrunk by 1-4% by volume, consistent with the early tests. Cross sections of the targets were cut and polished to examine the pellet/cladding gap; they were found to be small and within the design constraints. See Figure 5.

This set of irradiations provided the information needed to complete the pellet down select, verify dimensional changes and gas release, and confirm that the full length target design would meet the irradiation goals and comply with all the safety requirements. At this stage of testing, it was apparent that the maximum powder sintering temperature, the maximum pellet pressing force, and hydrostatic compression of the cladding on the pellet stack were all needed to create a high yield target that met the reactor safety requirements. Thus, only one pellet formula, the one with the highest ^{237}Np sintering temperature, remained for the full length target testing.

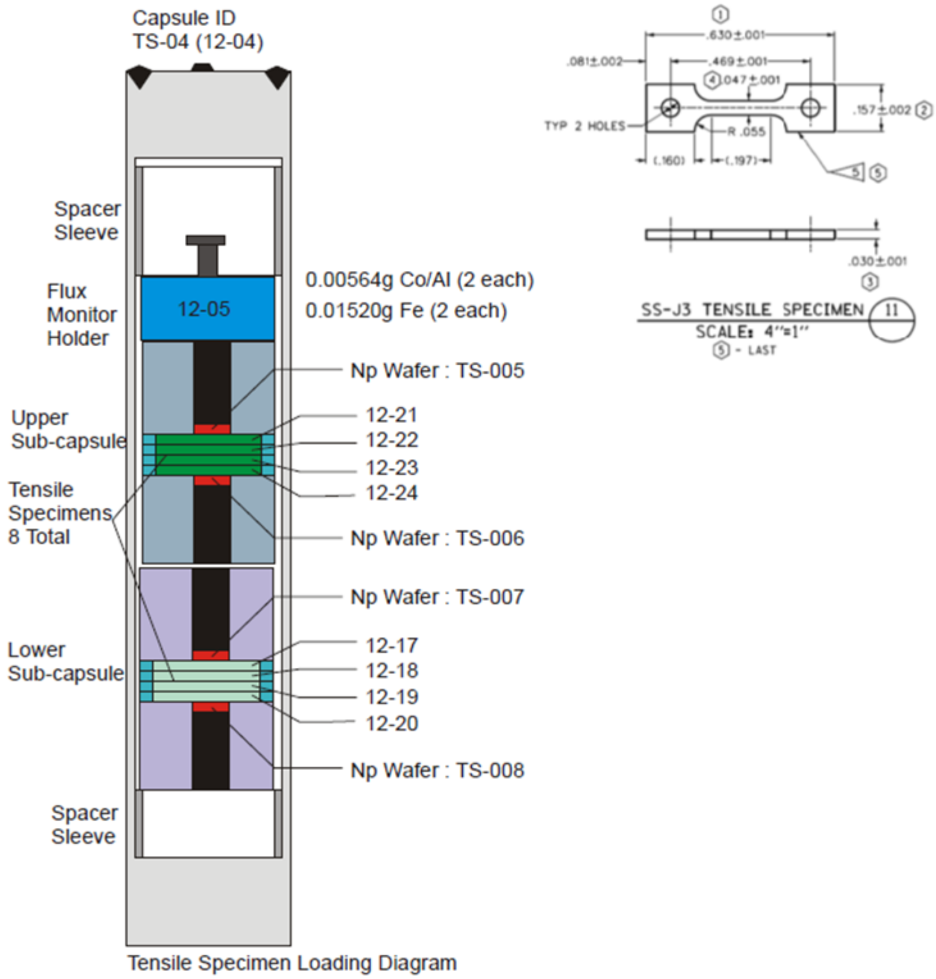

Figure 4. Special capsule designed for examining the radiation effects on the cladding material and the enclosed tensile specimens.

FULL LENGTH TARGET POST IRRADIATON EXAMINATION

A total 3 partial length targets underwent PIE. These were prototypical targets containing 50 pellets. These targets underwent the same examination as the previous targets with the addition of gamma scanning to determine the pre-PIE internal configuration.

The results of the gamma scanning are shown in Figure 6. As can be seen, considerable detail about the target internal structure can be determined. Small gaps between the pellets are acceptable; the major concern is that excessive pellet swelling can increase the pellet stack height and exert forces on the ends of the target. If these forces are high, target rupture is possible. However, in our case, the pellet irradiation has not gotten to the point where the pellet dimensions have exceeded their fabrication dimensions, thus for 2 cycles of irradiation swelling issues are not important. The two major issues for these targets are the point of maximum pellet shrinkage and the pellet gas release. The shrinkage opens the pellet/clad gap and the gas release adds low heat conductivity fission gases to the target internal environment. The combination of the two can increase the pellet temperature beyond acceptable levels. Dimensional data obtained on pellet diameter shrinkage and fission gas release for the full length targets is shown in Figure 7 and Figure 8. Fortunately, the design point and the selected pellet fabrication conditions result in acceptable performance with good safety margin for the general condition; the few outliners are under investigation.

Figure 5. Cross section of partial length target showing the pellet clad gap after irradiation (P-94, pellet #4, mount 6395).

CONCLUSIONS

A graded approach was under taken to recover the ^{237}Np target design for US production of ^{238}Pu based on past cermet experience. The approach included the irradiation of single pellet capsules to examine the behavior of the newly fabricated cermet pellets, followed by partial length targets to collect additional information in a nearly prototypical condition to allow for pellet fabrication technic down select, followed by a full length 2 cycle irradiation to complete the data collection and verify the target design under actual irradiation conditions.

The result was a set of fabrication conditions, a target design, supporting data, and the necessary safety documentation for a high yield target. The next step is to quantify the actual ^{238}Pu production rate.

ACKNOWLEDGMENTS

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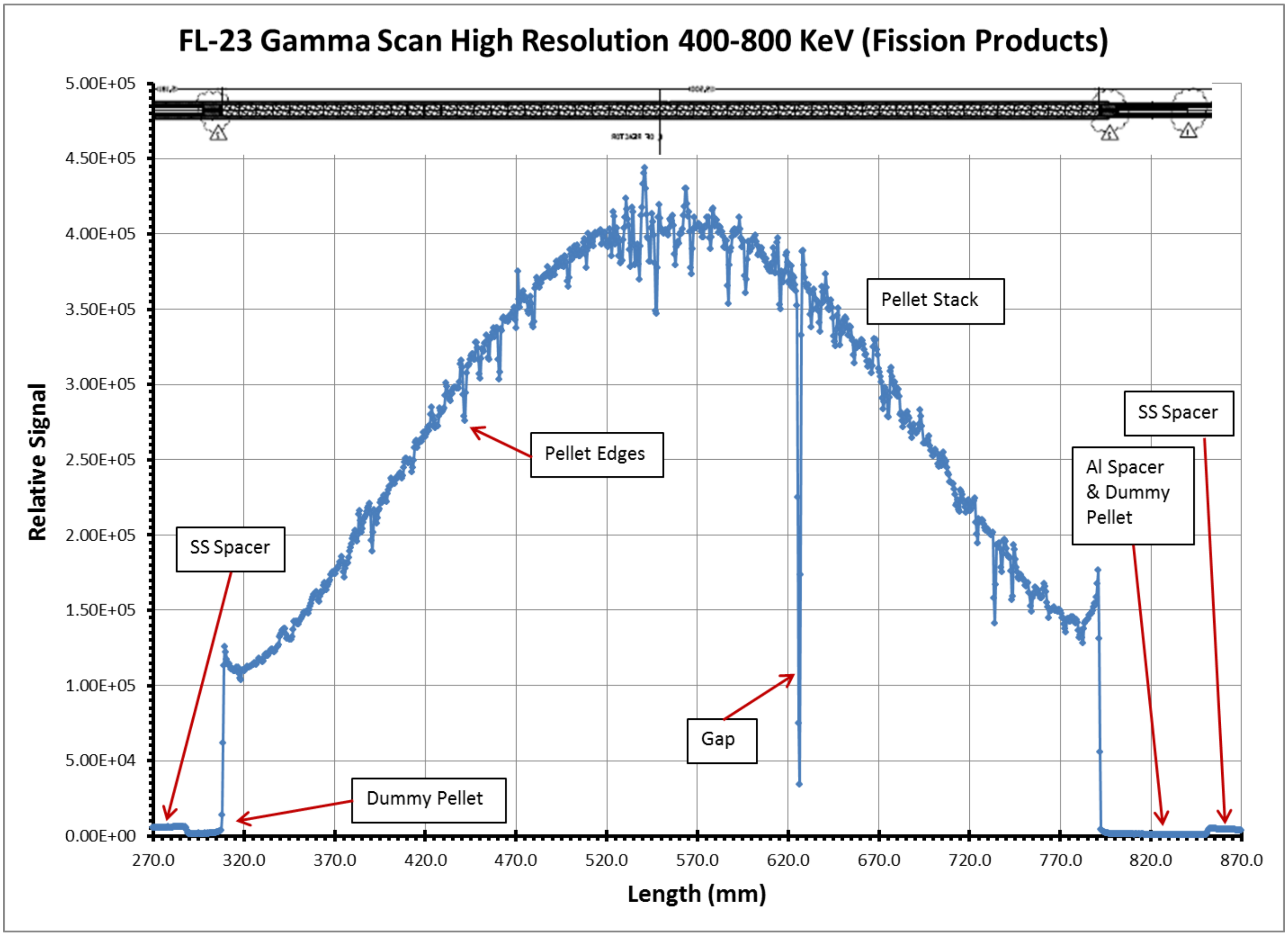

Figure 6. Results of gamma scanning a 2 cycle full length target.

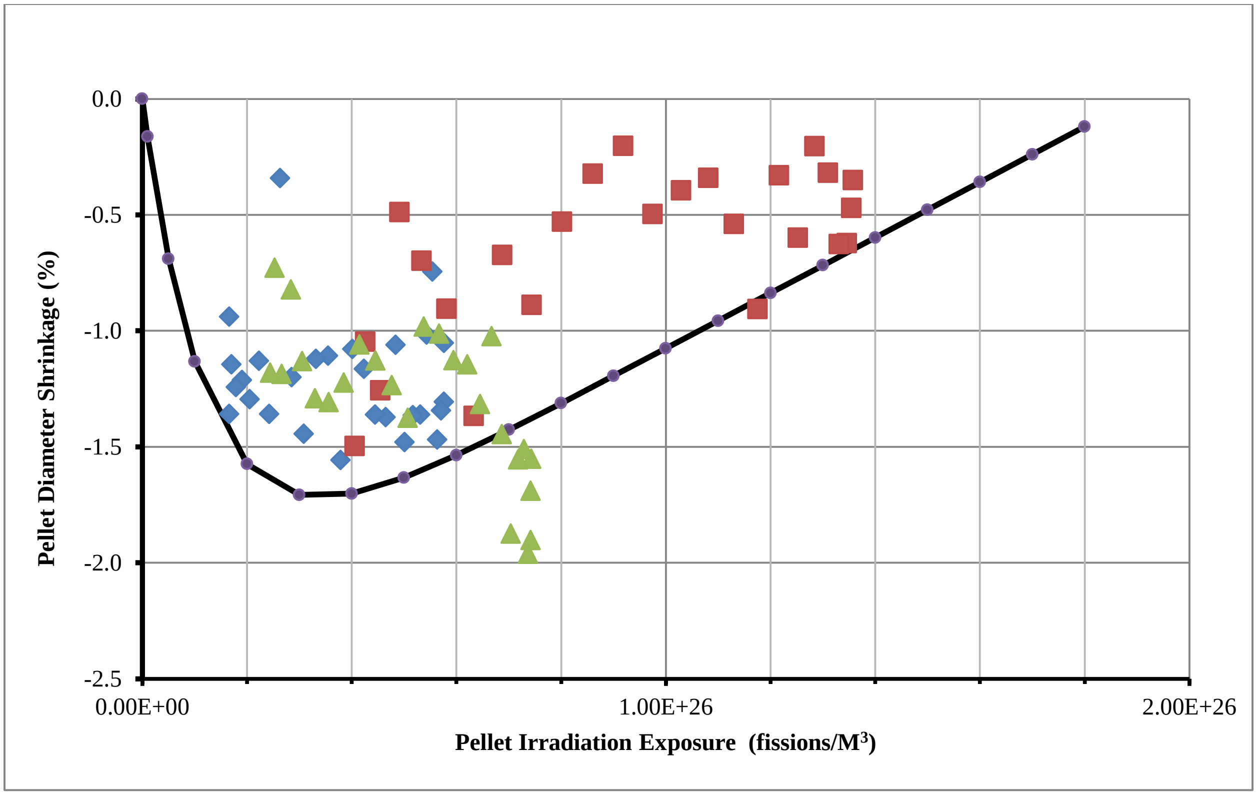

Figure 7. Pellet diameter shrinkage as a function of exposure. The solid line is the desired lower bound for a conservative safety analysis.

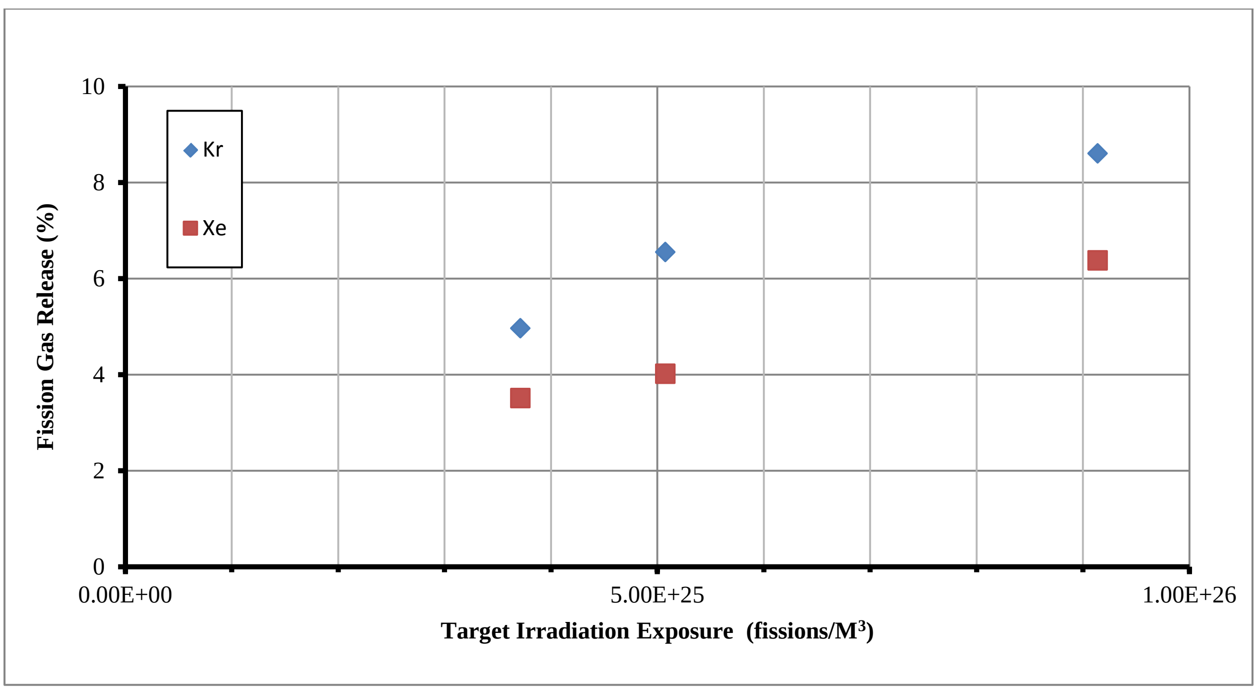

Figure 8. Fission gas release as a function of exposure.