

Supporting Information for:

Measuring Signatures of Fuel Irradiation in Large Particle Samples

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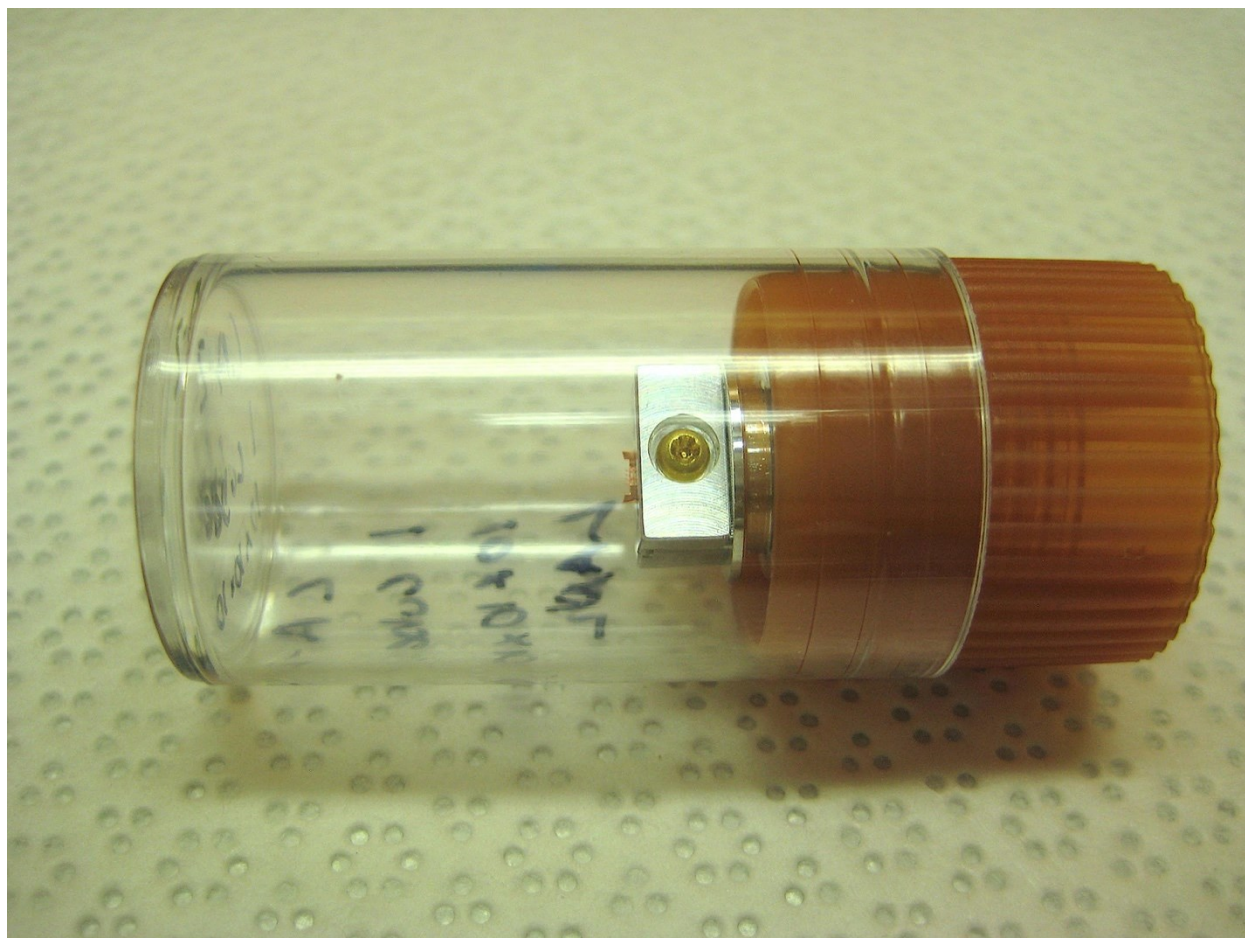


Figure S1. Photograph of a vial containing a FIB sample as received. The stainless steel rectangle on the bottom of the vial lid is the SEM stub. The FIB sample, too small to see with the naked eye, is mounted to the copper comb which is attached to the SEM stub.

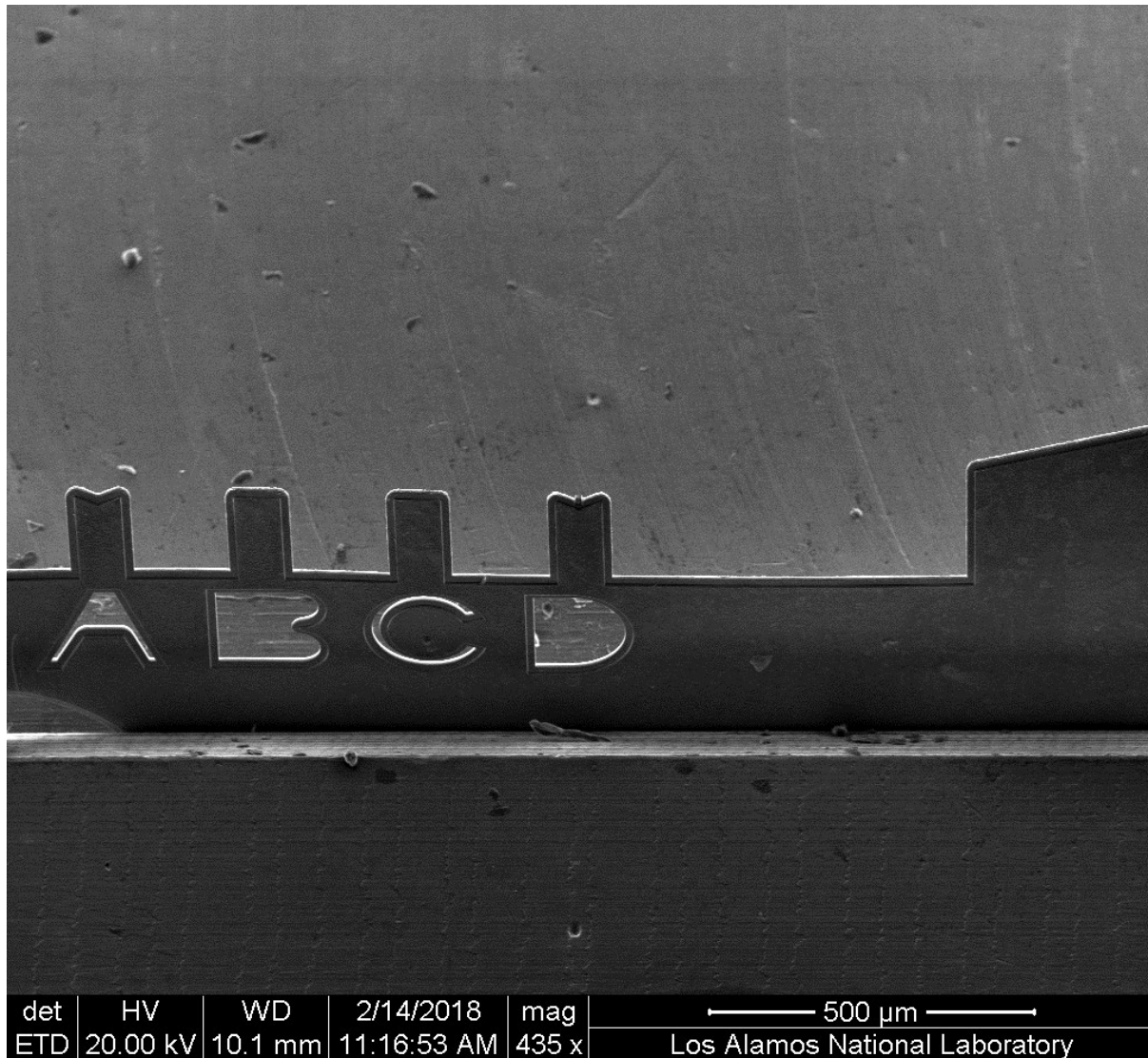


Figure S2. SEM image of a copper comb with a mounted FIB sample.

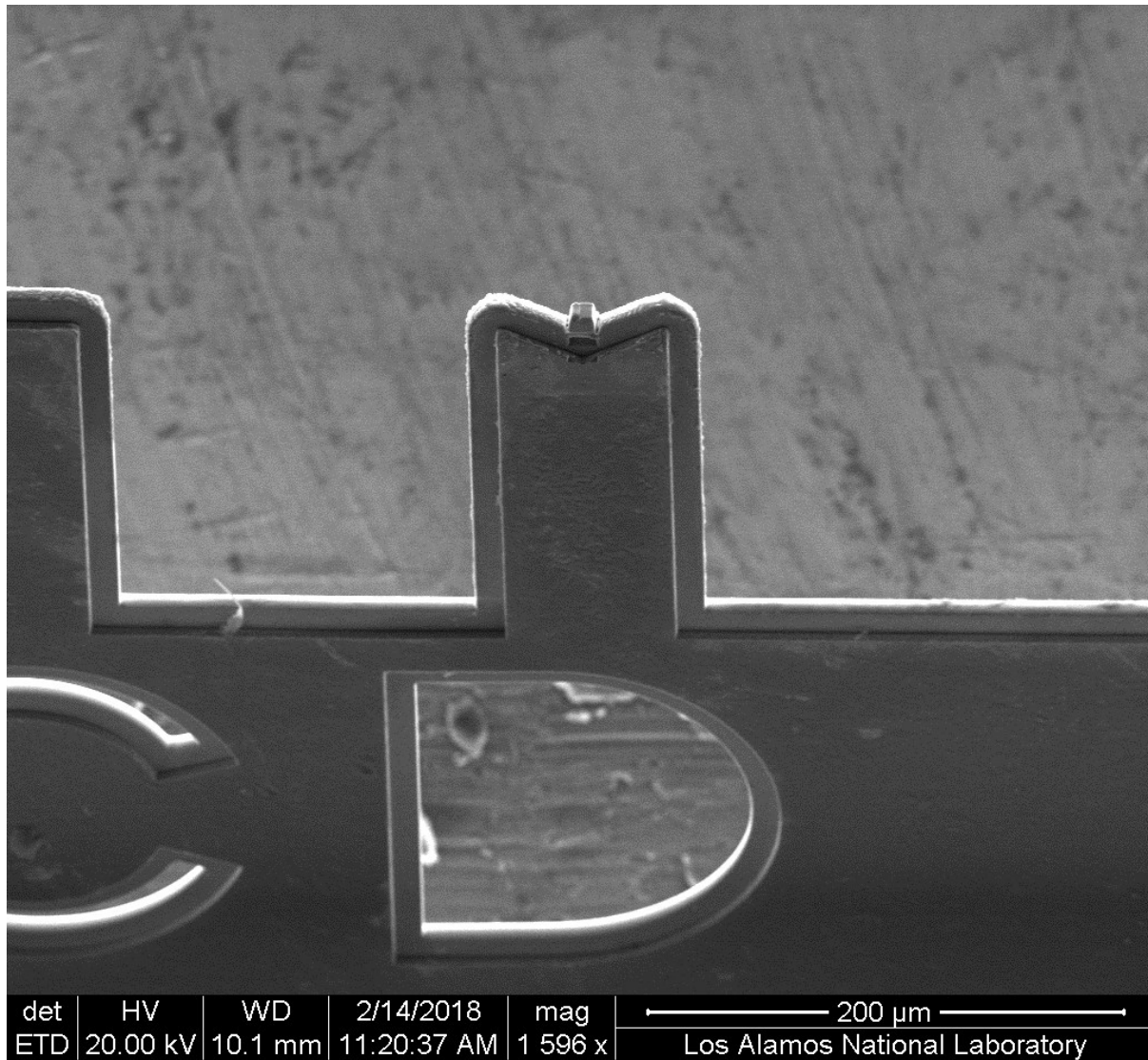


Figure S3. A closer view SEM image of a FIB sample mounted to a copper comb.

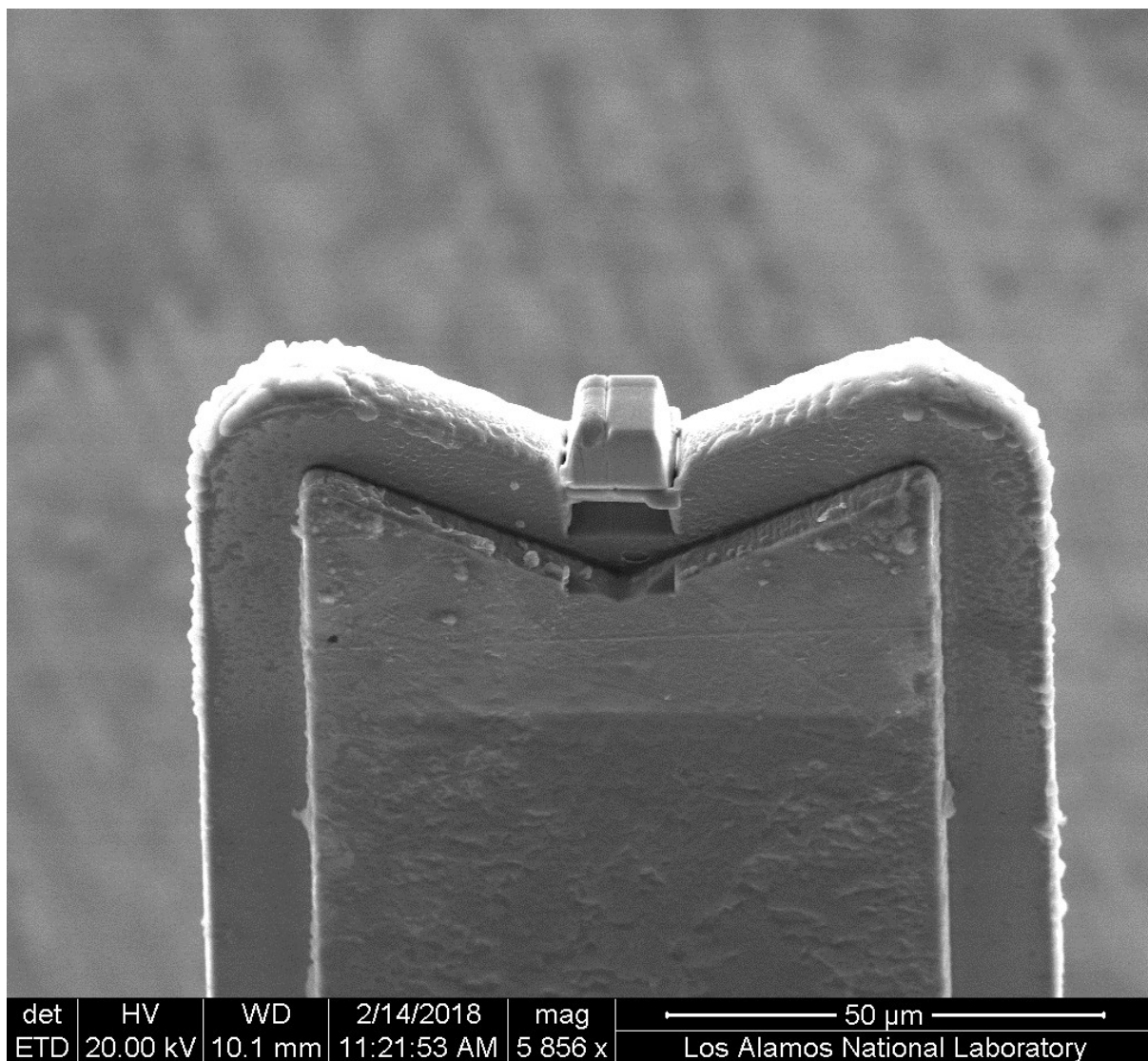


Figure S4. Close-up SEM image of one of the FIB samples mounted to a copper comb. A piece of the fuel is missing near the front top left corner of the cube. This FIB cube was cut in the same batch as the samples analyzed in this study, but was not destructively analyzed.

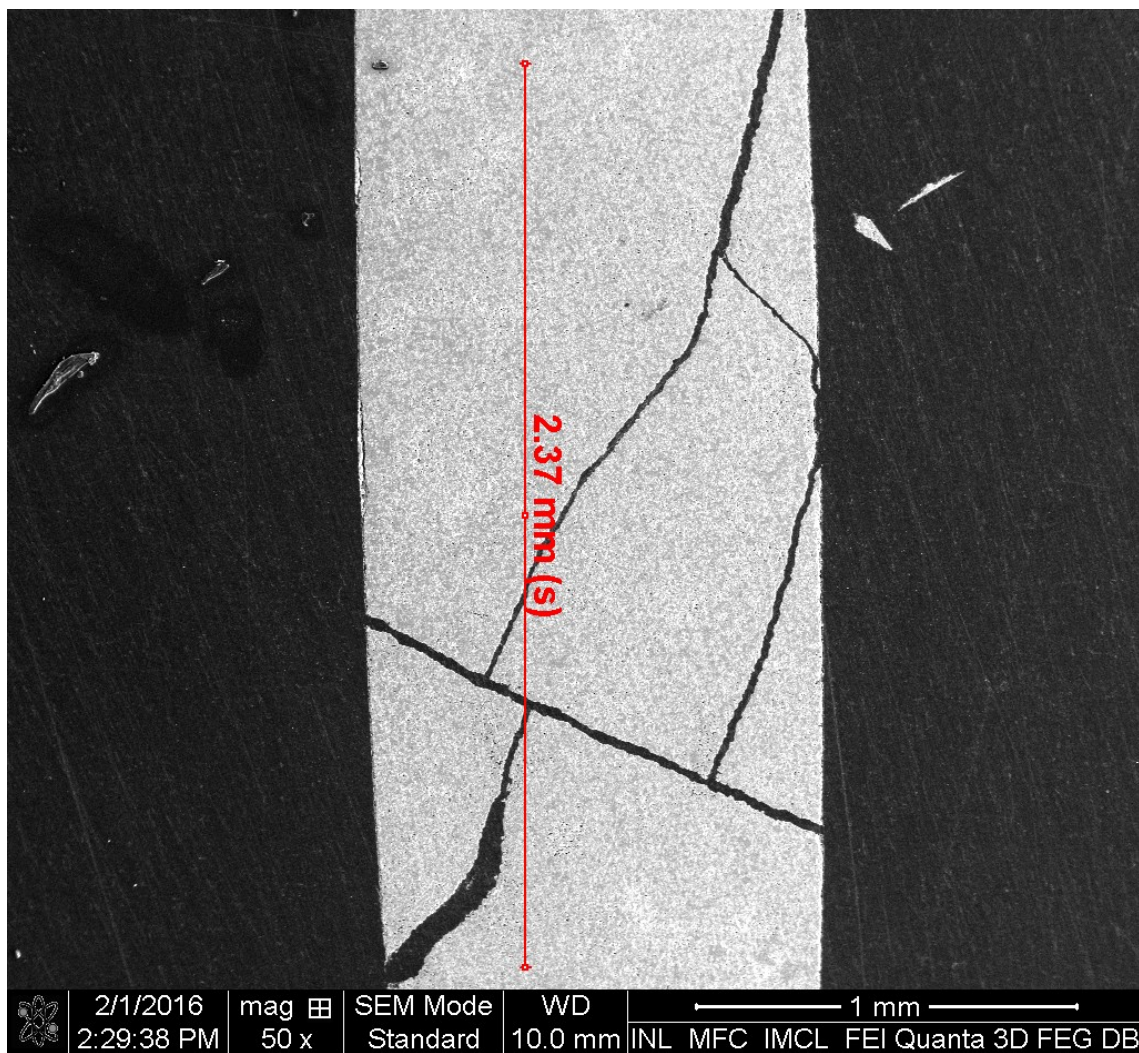


Figure S5. SEM image of a segment of the BR3 fuel at position 1, mounted in epoxy and used to cut the FIB samples. A considerable amount of cracking is evident; the fuel is known to be friable.

Note on the total amount of actinide measured in the FIB samples.

The FIB samples measured in this study were dissolved immediately upon opening of the vial and were not examined under the SEM. In the isotope dilution actinide analyses, some of the FIB samples (1-C1 and 3-B) contained less actinide than would be expected based on the original dimensions of the cube. For example, sample 1-C1 was found to contain 0.138(6) total ng U, whereas ca. 9-12 total ng U were measured in other FIB cubes of similar size in this study. Plutonium and americium measurements were correlated with the uranium in a comparable trend. It is possible that the lower amounts of actinides observed in these samples is due to loss of a part of the cube due to cracking or disintegration. Consistent with this idea, an SEM image of a related cube cut as part of the same batch (not analyzed as part of this work, Figure S4) showed a portion of the cube missing. The high-burnup BR3 fuel is known to be porous and

friable, and cracking was observed in the solid mounts used for cutting the FIB samples (Figure S5). Due to this known potential for cracking and sample loss, it was not possible to take SEM images of the samples that would be used for destructive analysis in order to minimize the amount of physical handling of the samples. As described in the text of the main paper, the sample vials were opened in a chemical hood and the copper comb and FIB sample were immediately dissolved.

In order to test whether the FIB sample had detached from the copper comb inside the vial during shipping, the vial was also rinsed multiple times with HNO_3 , which was collected in a clean Savillex vial. This rinse was then treated as an unknown sample and measured for total U content. The result of this measurement was ~ 1 pg total U, comparable to the process blank, indicating that the sample (nominally 10 ng U expected) was not in the vial either. We surmise that the sample was lost in handling prior to being placed in the vial for shipping.