DETERMINATION OF $^{235}$U IN HIGHLY ENRICHED REACTOR FUEL STICKS

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Measurement of uranium in highly enriched reactor fuel sticks has been achieved by elaboration of the method of Amiel. Calibration of the method was achieved by three methods: comparison of dissolved sample with aqueous standard, comparison of sample with handcrafted standard stick, and the extrapolated-aliquot procedure. Precision and accuracy of < 0.5%, relative are achieved in <2 min measurement time on a production basis. Precisions of <0.1% relative are achieved in replicate measurements.

Introduction

The measurement of $^{235}$U in reactor fuel sticks is an important task in the construction of Gulf General Atomic's HTGR reactor cores, which incorporate numerous such sticks of approximately 1.4 cm diameter by 5 cm length and over one hundred milligrams $^{235}$U content (actually from 105 mg to 255 mg $^{235}$U over all production types). Inasmuch as the exact uranium enrichment has been determined by mass spectrometry, it is possible to carry out the necessary measurements by determination of total uranium. However, the sticks are composed of highly refractory particles in which highly enriched uranium, as the carbide, is coated with graphite and silicon carbide. It is difficult and time consuming to thoroughly crush and extract the uranium from the samples for the more traditional methods of analysis. Therefore, an elaboration of the delayed-neutron method of Amiel for the neutron activation analysis (NAA) measurement of $^{235}$U was devised, and it has been successfully applied to the task at hand.

The delayed-neutron method is direct and rapid, since it basically requires only that the sample be irradiated with thermal neutrons to fission $^{235}$U and then counted with neutron detectors to measure neutron-emitting fission products. However, in the samples of interest significant self-shielding effects were present. As a consequence, application of the delayed-neutron method was carried out within the context of a difficult standards problem. The operations required to transform fuel sticks into simple solutions that might be directly comparable to primary standard solutions are regarded as potential sources of error. Similarly, the construction of
primary standards in the form of fuel sticks involves operational sequences that are potential sources of error. Thus, although both of these approaches were used, the method of varying sample size, with exploration of response vs sample weight to zero sample weight, was also used for standardization purposes, as will be described. This will be called the extrapolated-aliquot method. Upon achieving a consistent definition of response function by all three methods, which indicated a minimum chance of error, secondary standards sufficed to maintain accuracy of measurement.

Experimental

Apparatus and basic procedure

Samples and standards were contained in sealed polyethylene vials. They were irradiated by insertion through a pneumatic transfer system into the F-Ring of a TRIGA Mark I reactor. At the end of the irradiation, they were delivered directly to the counting station. Samples were processed sequentially.

Electronic timers were utilized to control the sequence of operations. As the sample entered the core of the reactor it passed a photo-sensor, which started one timer; as it returned from the core it was again sensed, which stopped the timer. At the same time another timer was started which controlled the delay time before starting the count and the duration of counting.

The counter was comprised of 6 high-pressure, 2" diameter by 12" long BF$_3$ tubes imbedded in a block of polyethylene such that their centerlines were 6.35 cm from the center of the sample annulus. Its efficiency for counting delayed neutrons was 7%.

In early experiments the experimental cycle consisted of a 45 sec irradiation, 40 sec delay, and 60 sec counting period. Later, the cycle was revised to 30, 30 and 50 sec, respectively, which enabled a faster throughput of samples without a significant sacrifice of precision.

Reactor power during irradiation was inversely proportional to the level of $^{235}$U in the sample, being 25 W for 100-400 mg of $^{235}$U, 250 W for 10-40 mg of $^{235}$U, etc. Thermal neutron flux at the sample position was $\sim 10^7$ n cm$^{-2}$ sec$^{-1}$ per W. Thus, $<10^{10}$ fissions occurred in any given sample irradiation.

Samples and standards

In addition to the fuel sticks described earlier, which were non-destructively analyzed, four aliquots, weighing on the order of 300, 500, 800, and 1000 mg, were prepared from each of several crushed and ground individual sticks. Also, in several instances sticks were crushed, ground, burned (to burn off graphite), re-ground, and dissolved to obtain aqueous solutions. The dissolution procedure did not dissolve the residual silicon carbide; thus the solution was filtered prior to