Discovery That Nuclear Fission Produces Tritium

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Abstract

Webster defines serendipitous as 'the finding of valuable or agreeable things not sought for'. This definition truly fits the surprising scientific discovery made early in the operation of the Savannah River Plant that tritium is a product of nuclear fission. Entirely by chance in 1957 in a hydrologic tracer study with tritium at a waste water disposal site, it was discovered that tritium was present in very measurable amounts in irradiated nuclear fuel. The tritium-in-fuel phenomenon remained unexplained for two years until (again by chance) it was proposed to apply its presence as an analytical tool to measure reactor exposure of nuclear fuel. This idea, which quickly turned out to be impractical, caught the attention of the research staff at what was then the Savannah River Laboratory and a basic scientific inquiry was eagerly encouraged. An all-out study was completed in the remarkable span of ten weeks, ending in the successful discovery of an entirely overlooked portion of basic nuclear data.

Introduction

With the advent of the huge research programs needed to develop the atomic bomb, chemists and physicists had a great time discovering and characterizing the hundreds of byproducts created in the process of nuclear fission. By the late 1940s, it was assumed that all radioactive fission products with half-lives greater than a few seconds had been identified. In these studies, attention focused logically on the products of binary fission: ternary fission was recognized as a curious source of helium as energetic alpha particles. A few studies sought other light particles without notable success. No speculation existed that tritium might be formed this way; however, if anyone had looked, not by examining individual fission events as a physicist would, but by looking at the accumulation of trillions of events, as a chemist would, it would have been an easy discovery. Tritium has a moderate half-life of 12 years, is easy to separate chemically from interferences, and can be measured precisely. By the late 1940s, research attention had shifted away from 'old hat' fission phenomena to the formation of new elements by neutron and charged particle addition.

Discovery of Tritium in Fuel Reprocessing Waste

At the beginning of operation of the Savannah River Plant's (SRP) two irradiated-fuel reprocessing plants, lightly contaminated condensate from waste evaporators was discharged into earthen seepage basins. The Health Physics Section (HP) was developing an understanding of the movement of these radioactive contaminants into the groundwater by monitoring a series of nearby wells. Monitoring data from the wells was pretty barren for ionic fission products because the soil, as expected, was sorbing these products. Henry Horton, a specialist in soil science, and myself (Ed Albenesius), an organic chemist, ran this program. We had a crude model of groundwater movement based on hydrology, but we were eager to confirm data in the field. So we came up with the bright idea of 'spiking' a well with a small amount of tritiated water that would move freely with the groundwater. We would then measure its arrival behavior at nearby downslope wells. We spiked a well in December 1956, and, to our surprise, a week later the first sample of the nearest downslope well, 20 feet away, contained tritium, a rate of movement 10 times what we

expected. To our complete astonishment, a hurried sample of a well 100 feet away also contained tritium! We then rushed back to the seepage basin and discovered that it was the tritium source; subsequent careful analysis of all the waste streams feeding the basins traced the source to the process vessel in which the uranium fuel was dissolved to start the separations process. The presence of tritium was also confirmed in the enriched uranium fuel waste.

Over the next two years, as we strove to absorb the significance of this discovery, the reality was that we now had an extraordinary tool to understand the hydrology of the seepage basin system. With a perfect tracer leading the way, numerous soil cores were taken to optimize the placement of monitoring wells and to develop seepage basin hydrology in three dimensions; these steps enabled credible projection of future basin performance. In all, over 200 monitoring wells were installed over the next 10 years.

Lost in the shuffle for the first two years was any effort to understand scientifically why the tritium was present in the irradiated fuel. Off-the-cuff speculation tended to dismiss its presence as possibly due to a lithium impurity in the fuel (lithium splits into tritium and helium in a nuclear reactor). Or the tritium diffused into the fuel from the reactor heavy water moderator, where it is an impurity that grows in by neutron capture in deuterium. Or perhaps it might be a contaminant related somehow to the large amounts of reactor irradiated lithium-aluminum target rods used in the Site's large-scale tritium production process. Surely, the casual thinking went, with all that tritium on the Site, it should not be too surprising to see some turn up in unexpected places such as waste basins. The turn to scientific investigation of the actual source of the tritium and the completion of this whole serendipitous process required one more unplanned event.

How the Scientific Study Began

Two years after the discovery of tritium in the fuel separations waste, I was transferred from the Health Physics Section to the Analytical Chemistry Division of the Technical Division, the research arm of the Site. The environmental chemistry emphasis of the old assignment instantly supported the process chemical research in the new assignment. An intriguing problem at the time was the measurement of burn up (consumption of nuclear fuel in various reactor configurations). Among existing methods of burn-up determination, isolating and analyzing a specific fission product, such as Cs-137, was most frequently employed. These methods were difficult analytically and yielded imprecise data.

At the time of the discovery of tritium in the waste from the fuel separations plants, calculations indicated a rough correlation of tritium quantity with reactor exposure of the fuel (about 1 tritium atom per 40,000 fissions). The idea now surfaced that if correlation could be proved and precisely measured, the tritium in nuclear fuel could offer a useful approach to determining burn up because, analytically, tritium (as tritiated water) is easy to isolate and precisely measure. I took my idea to Harold Kelley, my research manager, and proposed a feasibility study. Kelley's reaction was one of unbridled enthusiasm. Hearing of the tritium-fuel finding for the first time, Kelley sensed that a potential scientific discovery was on the doorstep. At Kelley's urging, I discussed the tritium data with one of the staff's reactor physicists (Dan St. John) seeking a clue that might link tritium with fission phenomena. St. John knew of no such link for tritium, but pointed out that the helium ion is known to occur in a rarer mode of fission known as ternary fission. This was an extremely valuable lead that provided a focus for thorough search of the scientific literature on fission phenomena.

Search of the Ternary Fission Literature

No report could be found in the literature citing evidence for or against the formation of tritium in the process of nuclear fission. The phenomenon of alpha particle emission in ternary fission had been extensively studied. The mechanism had been well established. No evidence was found for the formation of tritium or deuterium. However, in one study of the energy distribution of fission alpha particles, the appearance of tritons was noted but was dismissed as an inconsequential interference. Studies of proton (the third hydrogen isotope) formation in fission were carried out by several authors. Protons were observed in a study with photographic emulsions but were discounted as collision products. Protons were also observed at a frequency of 1 in 5000 fissions in another study with a carbon dioxide range chamber. However, a theoretical study stated that proton emission should be practically forbidden in comparison to alpha emission. Thus, the literature had no references on the formation of tritium in fission and had conflicting evidence for the proton. The conversation with St. John and the resulting focus of the literature review had now taken the study from concept to hypothesis. What remained to be done was to prove the hypothesis in the laboratory.

Proof that Nuclear Fission Produces Tritium

The laboratory experiments to eliminate speculation as to alternative non-fission sources of the tritium and, if successful, to measure precisely its yield in the fission process required the skills of an accomplished radiochemist. Such a person was Bob Ondrejcin, who stepped forward to design and conduct these exacting studies (Albenesius 1960).

Three experiments were carried out to provide proof that the radioactive species being measured was tritium, that the presence of tritium was not due to irradiation of a lithium impurity in the uranium fuel, and that the tritium did not come from the heavy water moderator of the reactor. The experiments were as follows:

- A sample of water from the acid solution in which irradiated uranium had been dissolved was converted to hydrogen, and the gas was diffused through a palladium barrier. The diffusate was reoxidized to water, and the expected tritium content was verified.
- The lithium content of the uranium fuel was determined. A sample of uranium from a typical fuel element was dissolved in nitric acid, and the uranium was precipitated. The supernate, which contained any possible lithium impurity in solution, was analyzed by emission spectrography. The lithium content was less than 6% of the amount required to produce the level of tritium observed in irradiated uranium.
- A sample of enriched uranium was irradiated in a graphite-moderated experimental reactor that contained no heavy water. The sample was dissolved in nitric acid, and the water was separated by distillation. The ratio of tritium to fissions was comparable to the ratio in fuel irradiated in heavy-water-moderated systems.

For the precise measurement of the yield of tritium in fission, eight sections were cut from rods of uranium irradiated in an experimental fuel assembly in an SRP production reactor. Total fissions in the assembly were calculated from measurement of coolant flow, temperature, and neutron flux. The cut sections were cleaned of any adherent film of tritiated deuterium oxide, then dissolved in nitric acid in an apparatus designed to collect the offgas. About 25% of the total tritium was found in the gas phase. The overall yield of tritium was calculated to be 1.05 +/- 0.09 atoms per 10,000 fissions.

Following publication of the discovery of tritium as a product of fission (Albenesius 1959), two independent studies in 1961 confirmed tritium emission in the spontaneous fission of californium-252 and measured its energy

distribution. These works also proved our hypothesis that the tritium was formed as the triton in ternary fission. Emission of the triton in the fission of uranium was also independently verified in 1963. The fission yield of tritium for uranium was also independently confirmed in 1962.

References

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Biographies

Edward L. Albenesius

Degrees:
College of Charleston 1944-47
AB Chemistry University of North Carolina 1947-51
Ph.D. Organic Chemistry
AEC Fellow 1949-51
Employment:
1951-89, E.I. du Pont de Nemours & Co. SRP/SRL
1989-92, Westinghouse Savannah River Co.
Career Highlights:
1951-53 Research Chemist, Health Physics SRP
1953-59 Supervisor, Environmental Monitoring and related programs
1959-64 Research Supervisor, Analytical Chemistry Division, SRL
1964-87 Research Manager, Managed a series of SRL programs in support of process development, environmental chemistry, and low-level and transuranic waste management.
1988-89 Assigned to DOE HQ; Coordinated revision of DOE Order 5820.2A Radioactive Waste Management
1989-92 Coordinated research and education initiatives for WSRC with the S.C. Universities Research and Education Foundation (SCUREF)
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1942-43 Research Chemist, Heavy Water Process Dev., Columbia Univ.
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1945-50 Research Chemist, Polychemicals, Experimental Station
1950-53 Research Chemist, Explosives Dept., AED, Wilmington, DE
1953-67 Research Manager, Analytical Chemistry Division, SRL
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Daniel S. St. John

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Career Highlights:
1944 Los Alamos (atom bomb components) U.S. Army
1949-50 Research Chemist, Grasselli, Cleveland, OH
1950-53 Reactor Physics training, Argonne National Laboratory, Chicago, IL
1953-58 Research Supervisor, Theoretical Physics Division, SRL
1958-64 Research Manager, Theoretical Physics Division
1964-70 Research Manager, optical and ultrasonic holography project, Development Department, Wilmington, DE
1970-85 Laboratory Director, Explosives Department, at the Experimental Station. During this period, the Explosives Department Laboratories were combined with Nylon research to form the laboratory for the new Petrochemicals Department. Research areas included simulations and reaction kinetics for the production of acrylonitrile and statistical analysis of stratospheric ozone measurements
1985 Departmental Research Fellow, Petrochemicals Department
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Employment:
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1989-92, Westinghouse Savannah River Co.
Career Highlights:
1951-52 Jr. Chemist, Argonne National Laboratory
1952-55 Chemist, Setup/Start-up analytical control labs, SRP
1955-59 Laboratory Supervisor, Plutonium Analysis, SRP
1959-65 Analytical Chemist, uranium chemistry research, SRL
1965-69 Senior Chemist, titanium corrosion research, SRL
1969-73 Research Chemist, waste tank corrosion research, SRL
1973-83 Staff Chemist, aluminum and steel corrosion studies, stress corrosion cracking research, SRL
1983-92 Research Staff Chemist, nuclear reactor vessel corrosion research, SRL/SRTC
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