LA-UR-89-1347

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TITLE NUCLEAR FISSION AND NUCLEAR SAFEGUARDS: COMMON TECHNOLOGIES AND CHALLENGES

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SUBMITTED TO Invited overview paper presented at the American Nuclear Society/National Institute of Standards and Technology Conference: "50 YEARS WITH NUCLEAR FISSION."

April 26, 1989 at National Academy of Sciences, Washington, D.C.

April 27-28, 1989, National Institute of Standards and Technology, Gaithersburg, Maryland.

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FORM NO. 836 R4

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NUCLEAR FISSION AND NUCLEAR SAFEGUARDS:

COMMON TECHNOLOGIES AND CHALLENGES

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ABSTRACT

Nuclear fission and nuclear safeguards have much in common, including the basic physical phenomena and technologies involved as well as the commitments and challenges posed by expanding nuclear programs in many countries around the world. The unique characteristics of the fission process--such as prompt and delayed neutron and gamma ray emmision--not only provide the means of sustaining and controlling the fission chain reaction, but also provide unique "signatures" that are essential to quantitative measurement and effective safeguarding of key nuclear materials (notably ^{239}Pu and ^{235}U) against theft, loss, or diversion. In this paper, we trace briefly the historical emergence of safeguards as an essential component of the expansion of the nuclear enterprise worldwide. We then survey the major categories of passive and active nondestructive assay techniques that are currently in use or under development for rapid, accurate measurement and verification of safeguarded nuclear materials in the many forms in which they occur throughout the nuclear fuel cycle.

INTRODUCTION

Nuclear fission and nuclear safeguards have much in common, from their roots in a common basic technology to the closely-coupled challenges of nuclear fission energy and the necessary assurances provided by nuclear safeguards. In keeping with the guidelines set by the organizers of this historic commemorative conference, the present review, which was requested to cover both history and contemporary developments, is presented in two major subject areas: the first sketches some first-hand perceptions and reflections on the history of nuclear fission and the subsequent emergence of nuclear safeguards, while the second subject area covers the present status and technical capabilities of modern nuclear safeguards. A common thread throughout the entire paper is the very epitome of this conference; i.e., the remarkable phenomenon of nuclear fission, and its practical application. The unique characteristics of the fission process and of fissionable materials that underlie the technology of all fission energy applications also provide the unique "signatures" that are essential to quantitative measurement and effective safeguarding of key fissionable materials against theft, loss or diversion. Through the years, fission energy and safeguards have been closely interrelated, not only technically, but also in other ways including certainly political, and this close coupling is ongoing today with effective and credible safeguards an indispensable component of a viable and expanding nuclear enterprise worldwide.

NUCLEAR FISSION --AN INDIVIDUAL HISTORICAL PERSPECTIVE

As a point of departure, we look back nearly a century to the year 1905, when Albert Einstein published his theory of relativity; ever since then it has been realized that, based on mass-energy equivalence (the celebrated $E = mc^2$--probably the world's most famous, and fabled, scientific equation ever) there was a theoretical possibility of releasing enormous energy from matter (and subsequently the "curve of nuclear binding energy" clearly confirmed this). As we now know, nuclear fission was actually first produced in 1934 by Enrico Fermi and his co-workers when they irradiated many elements including uranium, with the newly discovered neutrons. They found a number of different β-activities to be produced from uranium, but believed that these were due to neutron capture. Later radiochemical work indicated some of the new activities were from elements chemically similar to the much lighter elements Ba, La, etc.

Fission remained unrecognized until January 1939 when the meticulous work of the German radiochemists O. Hahn and F. Strassmann, showed that these products were not merely chemically similar to lighter elements, but were lighter

elements.[a] In that same month Lise Meitner and Otto Frisch (in the January 16 issue of Nature) named the new process "fission," predicted that the fragments should have large kinetic energies, and explained the process in terms of a liquid-drop model. Also in that same month of January 1939, Enrico Fermi arrived in the United States from Fascist Italy.

Following the discovery of fission, confirming experiments were quickly carried out in laboratories around the world. It was soon discovered that neutrons were produced in the fission process, and that almost all of the fission of uranium was occurring in the relatively rare isotope, ^{235}U.[b] In that same year (1939), Niels Bohr and John Wheeler published their theory of fission, based on the liquid-drop model, which is still fundamental to modern fission theory.

On the date of publication of the Bohr-Wheeler paper, September 1, 1939, Germany invaded Poland, the Second World War was underway, and fission suddenly took on a new importance. It was realized by many that a fission chain reaction might be possible, resulting in the release of very large amounts of energy. Thus, informed scientists were already aware in 1939 that it might well be possible to produce the destructive effect of many thousands of tons of high explosive with a single bomb containing a relatively small amount of fissionable material. It seemed probable that Germany would press ahead with this development. Apprehensive of this possibility, scientists in the rest of the world largely ceased publishing fission results by 1940.

Work on fission was continued quietly at an increasing rate. In June 1942, the Manhattan Project was secretly undertaken in the United States, with the objective of producing nuclear weapons, if possible. Amazingly soon thereafter, the world's first self-sustaining fission chain reaction (lasting 28 minutes) was produced on December 2, 1942, under the direction of Enrico Fermi, the 'Italian Navigator," who with his co-workers had carefully assembled blocks of ordinary uranium and extra-pure graphite (as a neutron "moderator" to slow down the neutrons and increase the likelihood of fission) to produce a nuclear reactor, under the West Stands of Stagg Field Stadium at the University of Chicago. An incredibly short time later the gigantic gaseous diffusion facility was undertaken (starting in 1943) at Oak Ridge, Tennessee to separate the more fissionable isotope ^{235}U from ordinary uranium (a prodigious task that, it was later learned, had been considered a practical impossiblity by both Germany and Japan, at least in time for use in World War II).

A second candidate material for a self-sustaining fission chain reaction, and a possible nuclear weapon, was the plutonium isotope, ^{239}Pu, that had only recently been discovered by Glenn Seaborg and co-workers at Berkeley in 1941. It was thought, and later proved, that plutonium (and more specifically the isotope ^{239}Pu) should be fissionable by slow neutrons in the same way as ^{235}U. Following active research on the chemistry of plutonium at Berkeley in 1941 and early 1942, the Plutonium Project was established in early 1942 at the University of Chicago, and on August 20, 1942 the first chemical compound of plutonium, a fluoride containing only ~1 microgram of ^{239}Pu, was isolated. Just three weeks later the first actual weighing of a pure chemical compound of plutonium, PuO_2, took place on Thursday, September 10, 1942 at the newly-established University of Chicago Metallurgical Laboratory.[a]

It seems no great exaggeration to say that nuclear fission and the ensuing nuclear age have impacted, to a greater or lesser degree, the lives of most of the five billion inhabitants of this planet -- and in my own case this has been

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[a] Viewed from the historical perspective, it is most interesting that today, a half century later, we have the startling announcement/claim of a radically new, allegedly-nuclear, phenomenon popularly known as "cold fusion." Experiments to date are inconclusive (some are even disputed), and the verdict is clearly still out on "cold fusion"; at this writing the claimed large energy release from cold fusion is not understood--with conjectures ranging from the unprecedented possibility of nuclear reactions resulting from some type of chemical/molecular interactions to the very remote possibility of a radically new genre of ultra-high exothermic chemical reaction(s). Whether boner or breakthrough, the aroused scientific community will see to it that this issue is objectively and definitively resolved, and that Nature's truth will out!

[b] ALFRED O.C. NIER[1] at the University of Minnesota (per direct request of Enrico Fermi) separated a sample of ^{235}U using his recently developed mass spectrometer. The sample was then sent to Columbia University where it was used to confirm the rare isotope's responsibility for slow-neutron fission.

[a] It should be borne in mind that, because nuclear safeguards are directly concerned with fissionable materials and their potential for diversion and misuse, emphasis in this historical perspective is on sensitive fissionable materials (notably ^{235}U and ^{239}Pu --the critical components in nuclear explosive devices), how they were first obtained, and how they are used, safeguarded and controlled.

true "in spades."[3] On the same day that (plutonium) chemistry history was being made,--Thursday, September 10, 1942-- a wide-eyed young freshman arrived at the University of Chicago full of enthusiasm to pursue his intended major in the exciting field of chemistry! My assigned dormitory room on the 3rd floor of Hitchcock Hall at the corner of 57th and Ellis, looked directly over the West Stands of the football stadium at Stagg Field. During the course of that very busy and stimulating fall at the University of Chicago, I and many other young would-be scientists found the entire atmosphere electric and highly stimulating. Particularly impressive to many of us was a sealed-off heavily guarded area posted with the stern warning "U.S. Government Metallurgical Project--Keep Out." As we regularly passed by this area on the way to our Monday-Wednesday-Friday freshman calisthenics class, we would occasionally pick up black dust (Fermi's graphite) on the soles of our tennis shoes. Needless to say, we were totally oblivious to the history "the Italian Navigator" was making under our very noses; many on campus sensed that something really big and important must be going on, and we wondered when, and if, we'd one day find out what it was all about. That day did come nearly three years later on August 6, 1945, at Columbia University in New York City.

In 1943, I was recruited into the Navy "V-12" College Training Program and was sent to MIT in Cambridge, Massachusetts on the condition that I change my major to physics, and subsequently serve as a Radar Officer in the U.S. Navy. Thus I switched from chemistry to physics, and upon graduation from MIT in June 1945, all of us in the Navy V-12 program were immediately sent to Columbia for an intensive 90 day officer training course (the regular Navy derisively dubbed us "90-day wonders"). With Hitler's Germany just defeated, in May 1945, the war focus was now riveted on the far east, and we were being prepared for immediate sea duty and the coming massive ("million-man") invasion of Japan. Everyone understood that a U.S. invasion of the Japanese homeland would be tremendously costly in lives and resources, but it was to be the final big push that would end World War II and at last bring "peace to the world." In early Summer, 1945, however, the war was still raging, and though we didn't talk about it (the invasion) much, we all knew full well what was at stake for our country, the world, and also for each one of us individually.

Then on the afternoon of August 6, 1945, as we were marching from drill back to our training ship "The Prairie State," anchored in the Hudson near George Washington bridge, we heard the newsboys shouting "Truman announces revolutionary new atomic bomb dropped on Japan." Two burning questions rushed immediately into my mind: (1) might this end the war quickly, scratch the U.S. invasion, and save untold lives --both American and Japanese, including perhaps my own? and (2) could this have been what the super secret "Metallurgical Project" at Chicago was all about? Both of these questions were soon to be answered, in the affirmative. On August 15 the Japanese surrendered, the mobilization of the massive United States invasion force was called off, and World War II was over. On that unforgettable day, August 15, 1945, I found myself caught up in the wild, tumultuous throng that jammed into Times Square in Manhattan to celebrate "V-J Day" (Victory over Japan Day), and the return of peace, at last, to the world.[4]

Immediately after the war, my service commitment to the U.S. Navy was fulfilled with a year of sea duty ("magic carpet" duty, bringing troops back home from the far east) as a Radar Officer aboard the USS Kingsbury (which we irreverently dubbed the "dingleberry"). I then returned to civilian life to complete my graduate studies in physics (the Navy-imposed switch from chemistry had "taken"!), and September 1949 found me, or rather us (I now had a wife and newborn son), heading West in our "new" 1939 Ford Coupe to the University of California, Berkeley to do research under Professor Emilio Segre as a U.S. Atomic Energy Commission Postdoctoral Fellow. Berkeley was an extremely exciting place to live and do physics in 1949. E. O. Lawrence was Director of the University of California Radiation Laboratory and I was privileged to get to know and to work along side many leading scientists, several of whom had worked at Los Alamos in the Manhattan Project--people like Emilio Segre, Luis Alvarez, Owen Chamberlain, Robert Serber, Edwin McMillan, Larry Johnston, Hugh Bradner, and many others. From a technical and professional standpoint, I came to have a new appreciation and high regard for the quality and quantity of technical work that had been accomplished in such an incredibly short time at Los Alamos. I must candidly admit right here that I had already fallen in love with New Mexico on earlier visits to the Southwest dating as far back as 1939 --when J. Robert Oppenheimer himself was still exploring the strange and wonderful natural beauty of "The Land of Enchantment." Thus, when in the fall of 1951 I was offered a research staff position at

[3] As explained at the outset, this historical sketch is purposely written from an individual perspective (my own), a point that should be kept in mind, particularly in some of the recounting of personal impressions and recollections.

[4] It seems appropriate here to cite once again the earlier advisory footnote concerning the rather personal, first-hand-account nature of some of the reflections recounted here.

Los Alamos, I accepted, we arrived bag and baggage on "the hill" on a New Mexico star-studded New Years Eve, December 31, 1951.

I joined the staff of the Los Alamos Critical Experiments Group, under the leadership of H. C. Paxton, which carried out research on critical and subcritical assemblies of sensitive fissionable materials at the Pajarito Canyon Site some seven miles southeast of the Los Alamos townsite. The Pajarito Canyon site was chosen for critical assembly work so that its isolation could protect others from radiation if a criticality accident should occur.[a] Originally, critical assemblies (e.g., of ^{235}U and ^{239}Pu in various bare and reflected configurations) were employed specifically for the nuclear weapons development effort. Later, many experiments were aimed at establishing a wide range of criticality-safety guidelines and restrictions. Also, extensive investigations with various configurations of fissile and fertile materials in bare and reflected geometries were carried out to gain a better understanding of the physics of nuclear reactions in fissionable materials and of the fission process itself. Such investigations have included detailed studies of fission-chain dynamics and prompt neutron behavior of near-critical systems; e.g., "Rossi-α" measurements of prompt neutron periods in metal critical assemblies,[3] which provide an independent method of precise reactivity calibration. Clearly also of fundamental importance to reactor kinetics and precision reactivity determination are the prompt and delayed neutron and gamma ray emissions from fission, as discussed further below.

The use of intense prompt neutron bursts from the bare ^{235}U assembly, "Lady Godiva" (she's unclad), to study the detailed characteristics (e.g., group periods and yields) of delayed neutrons from fission provides one of many examples of the use of critical assemblies in nuclear fission research. The decay-group periods and yields of delayed neutrons are of particular importance in the control of nuclear reactors because they limit, or "pace," the rate of increase or decrease at which a fission chain reaction can proceed. Delayed neutron characteristics are measured by irradiating a small sample of fissionable material with a burst of neutrons (either high-energy "fast" fission neutrons or low-energy "thermal" neutrons), then rapidly transferring the sample to a shielded neutron detector and measuring the decay of delayed neutron intensity with time. Computer analysis into exponential decay-group yields and periods (half lives) is then performed by an appropriate iterative least squares fitting procedure. The ultra-high neutron intensities obtainable from a critical assembly such as Lady Godiva provide a unique and ideal irradiation source for such measurements. Figure 1 shows the Lady Godiva bare ^{235}U critical assembly (53 kg of 93% enriched ^{235}U metal) that was used for the extensive delayed neutron studies that were carried out in the mid-1950s at the Critical Experiments Facility at Los Alamos. For operation (always by remote control), the upper cap of the Lady Godiva assembly (see Fig. 1) was lowered, and the bottom cap was raised, to form a

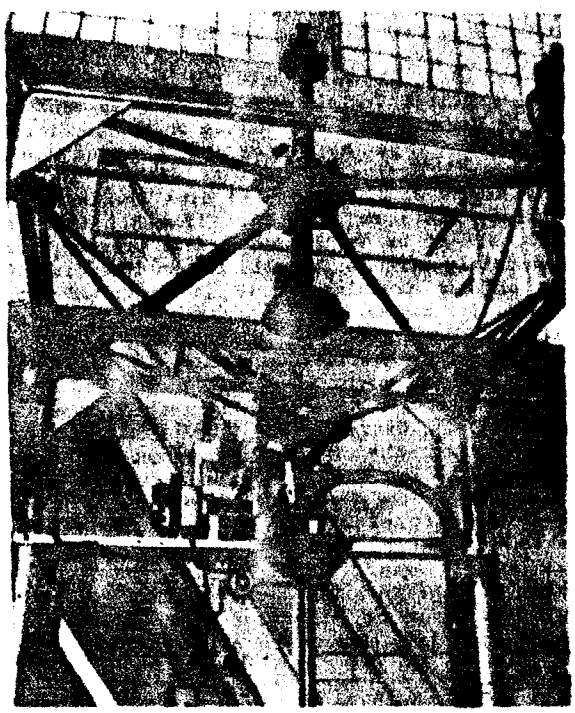

Figure 1. The Lady Godiva bare ^{235}U critical assembly (53 kg of 93% enriched ^{235}U metal) that was used at Pajarito site, Los Alamos to measure detailed characteristics of delayed neutrons from fission of various uranium, plutonium and thorium isotopes. For operation, the upper cap of Lady Godiva is lowered and the bottom cap is raised to form a near-sphere of ^{235}U without reflector.

[a] The first critical assemblies were manipulated by hand, a hazardous procedure that had led to the death of Harry Daghlian in 1945 and of Louis Slotin in 1946.[2] In each case the critical assembly being manipulated consisted of two mating Pu hemispheres (the "PLUTEX" core discussed below) surrounded by a neutron reflector (of tungsten carbide and beryllium, respectively). And in each case a component of the critical assembly had slipped into a more reactive position, producing a superprompt-critical pulse of radiation. The second accident led to a formal ban on hand operation, and a remote-control Critical Experiments Facility (with 1/4-mile separation between critical assembly laboratory and remote control center) was specially designed and rushed to completion at Pajarito Canyon site in 1947.

near sphere with essentially no reflector. The central section contained a diametral experimental access hole ("glory hole") with fillers and channels for two ^{235}U (93% enriched) control rods. Compensation for openings in the glory hole was provided by mass adjustment buttons of ^{235}U (93%) that fit into recesses in the top and bottom spherical caps. The glory hole was fitted with a 1/4-in. diametral transfer tube for rapid (50-msec) pneumatic transfer of the sample under study from its point of irradiation in the center of Lady Godiva to a 4π-shielded neutron counting geometry. In all, the Lady Godiva critical assembly was used to measure delayed neutrons from fast fission of ^{235}U, ^{233}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{232}Th, and from thermal fission of ^{235}U, ^{233}U and ^{239}Pu.[4]

Delayed neutrons as well as prompt neutrons and delayed and prompt gamma rays, are only a few of many basic fission characteristics that are important to the design, as well as efficient and safe operation of nuclear chain reacting systems of various types. However, this very broad topic would carry us far afield from the scope and thrust of this paper; for the present purpose, suffice it to say that unique, characteristic fission phenomena (such as fission neutron and gamma-ray emission) that are so important in sustaining and controlling the fission chain reaction also provide, by their very uniqueness, the characteristic "signatures" that are essential to effective measurement, safeguards, and control of sensitive nuclear materials. We shall return to this important topic presently in our discussion of nuclear safeguards.

Over the years the Los Alamos Critical Experiments Facility has worked with a great variety of fissionable material types, compositions and configurations, and the broad scope of this work at Pajarito Site has been well documented in two recent historical reviews.[5] For me, one of the most unforgettable and impressive experiences during some 13 years at Pajarito involved a sphere of metallic plutonium which we shall designate as plutonium core "X" or simply "PLUTEX." The PLUTEX core[6] consisted of two mating hemispheres (~9 1-cm diameter) of nickel-clad plutonium, each weighing approximately 3 1 kg. The gamma-ray and spontaneous fission neutron emission rates from PLUTEX were sufficiently low that the two hemispheres could be held in one's hands (i.e., together, as a sphere), which I did. The hemispheres were quite warm to the touch due to alpha-particle heating within the plutonium mass, as one holds them, the neutron monitor counting rate slowly rises due to the higher neutron multiplication rate produced by the neutron reflection and moderating (slowing-down) effect of the human hand (mostly H-O) around the core. What left an indelible impression on me in those early days was that this small grapefruit-sized metal ball that I could hold in my hands held the energy equivalent of tens of thousands of tons of TNT -due, of course, to the enormous energy release (~200 MeV/fission) in the process of nuclear fission, whose discovery we commemorate in this historic conference. Furthermore, for all its enormous destructive potential, this small rather benign-looking metal ball, besides being extremely valuable, was obviously very susceptible to concealment, theft, loss (it can happen!), and diversion.

The PLUTEX core is seen in an early (1946) photo (Fig. 2) as two bright metal (Ni-clad) hemispheres[a] placed side by side, flat side

[a] The spherical configurations that are so often characteristic of critical assemblies stem from very straightforward geometrical considerations of neutron "economy"; i.e., neutron production vs. loss. Neutron production is a volume effect, while neutron loss (primarily leakage, especially in bare systems) is a surface effect. The geometric shape that maximizes volume to surface is a sphere; therefore, for any given type of appropriate fissionable material, the minimum amount of (bare) material required to sustain a chain reaction is a sphere consisting of the (bare) "critical mass" of that material.

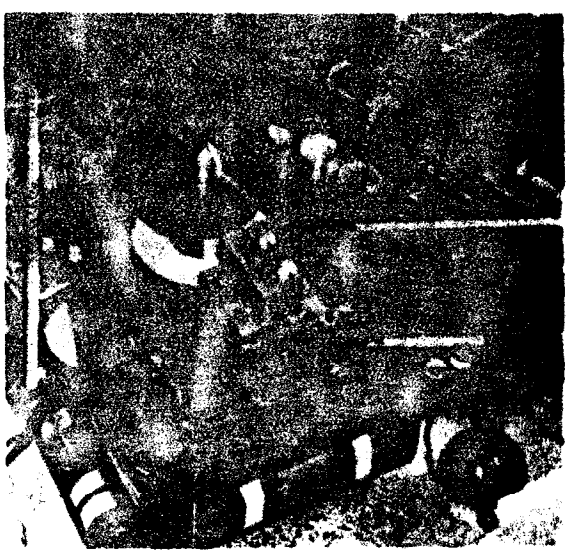

Figure 2. In this early (1946) Los Alamos photo the PLUTEX plutonium core is seen in the center of the photo as two bright metal hemispheres placed side by side, flat side down. Immediately behind the two PLUTEX hemispheres is part (the lower half) of a beryllium reflector assembly (consisting of a series of concentric nesting shells) that was used, with the core at the center, to achieve various core-reflector configurations.

[illegible] on stacked lead bricks. Immediately behind the two core hemispheres is a Be reflector (lower half-sphere) consisting of a series of concentric hemispherical shells complete out to a diameter of ~13 inches. In making the critical assembly, the plutonium core would be positioned in the center of the lower Be reflector and then enclosed within a succession of nesting upper Be hemispherical shells. The smallest upper Be shell (~4.5 in. O.D. and ~0.5 in. thick) is shown in position in Fig. 2. The largest upper Be hemispherical shell (~9.0 in. O.D.) is seen at the lower right under the soldering iron cord--to the far right of the Coke bottle, that is!). With the core positioned in the lower Be reflector, and the upper series of Be shells complete out to a diameter of 9 in., the assembly would become critical. In 1946 hand manipulation of such critical assemblies ceased completely, and all sensitive critical assembly operations have since been carried out by remote control (from a distance of 1/4 mile). The Flattop (19-in.-diameter "infinite" natural uranium reflector) assembly at the Critical Experiments Facility at Pajarito Site, Los Alamos (see Fig. 3) exemplifies modern specially-designed equipment for fully-remote-control critical assembly operations with sensitive fissionable materials. The striking contrast between Figures 2 and 3 underscores, perhaps more effectively than words ever could, the impressive progress made since the early days of hand operated critical assemblies; i.e, as regards both the mechanics of manipulation and operational implications.

Viewed from the perspective of nuclear safeguards today, my early years at Los Alamos working in the critical assemblies group provided invaluable hands-on experience and insight into the unique characteristics and behavior of both small and large quantitites of nuclear materials of various types, compositions and configurations. As a very sobering case in point, it was PLUTEX that drove home to me, as much as anything could, the vital importance of stringent accountability and controls over sensitive nuclear materials in the rapidly oncoming nuclear age --an issue (both technical and political) of steadily increasing worldwide concern over the years, that was to evolve eventually into the broad discipline now known as "nuclear safeguards."

EMERGENCE OF NUCLEAR SAFEGUARDS A PARTICIPANT'S PERSPECTIVE.

Viewed from a broad historical perspective, the dangers and potential for misuse of nuclear fission energy have been recognized since the dawn of the nuclear age, and by the close of World War II, some statesmen held the glimmering hope that placing all nuclear activities under international ownership and management could provide a basis for preventing, or at least

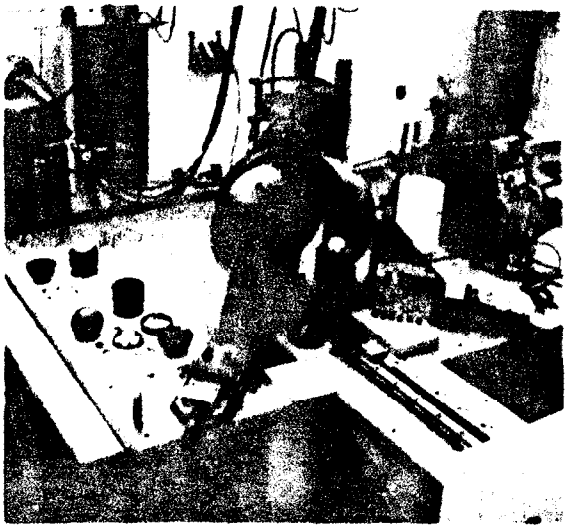

Figure 3. The Flattop critical assembly at Pajarito site, Los Alamos. A plutonium core (with bright nickel cladding) is shown in place on the central pedestal. Additional central cores of ^{235}U (93% enriched) and ^{233}U are displayed at the left with adapters and mass-adjustment inserts. For (remote) operation, with adapter in place, the pedestal is retracted into the stationary reflector hemisphere, and the two reflector quadrants (on ways at 45°) are moved inward to complete the 19" diameter spherical reflector of natural U.

restraining, the proliferation of nuclear weapons. In 1946, the Baruch plan proposed the creation of an international atomic development authority, to be entrusted with all phases of the development, use, inspection, and control of nuclear energy. The plan delineated the need for restraint in nuclear-weapon development and for international safeguards and penalties to prevent diversion of nuclear materials from civilian nuclear power programs. It also proposed that all nations forego the production and possession of nuclear weapons. Although many elements of the Baruch plan were eventually incorporated into international safeguards, in its time the plan was rejected, and by 1952 three nations had produced nuclear weapons. Secrecy became the fundamental nuclear policy of the United States and other nations. By the early 1950's many nations were seeking ways to acquire the benefits of nuclear technology and to develop their own nuclear energy programs. This burgeoning activity had an inherent potential not only for peaceful uses but also for military applications. The situation clearly called for renewed attempts to arrive at some form of international understanding, consensus, and constraint

President Eisenhower's 1953 proposal, the widely-hailed "Atoms for Peace" program, marked a fundamental change in U.S. nuclear policy. The program was designed to promote

international cooperation in the peaceful uses of nuclear energy and, at the same time, to establish international controls to ensure that the products of this cooperation would not be diverted to military uses.

A major event early-on in the Atoms for Peace program was the first United Nations Conference on the Peaceful Uses of Atomic Energy, this unprecedented worldwide conference was convened in September, 1955 in Geneva, Switzerland. As proclaimed by the vice president of the Geneva conference, Nobel Laureate I.I. Rabi, here for the first time scientists from the West and the East, and from around the globe were assembled to discuss the technical problems and challenges of the exciting new nuclear age. Insofar as possible, at the Geneva Conference all papers from various countries in a given subject area were grouped together in the same technical session or series of sessions. This format was used throughout the conference to facilitate detailed international intercomparison of new data and techniques. Thus the U.S. paper on delayed neutron measurements, which I was privileged to present, based largely on our recent work using Lady Godiva at Los Alamos, was directly followed in the same session by a Russian paper, also on delayed neutron measurements that had recently been carried out in the Soviet Union. Like all participants, I found this first truly international nuclear conference, providing for one-on-one interactions among scientists from around the globe working in the same field or specialty, to be extremely stimulating, significant, and memorable.[a]

I recall clearly that many of us in the U.S. delegation to the first Geneva Conference were filled with a sense of history, and some amazement too, at the open reporting of previously restricted information on fission data, fuel-cycle processes, and plant operations. Nearly every day, after late-night meetings of the U.S. delegation at the headquarters Hotel du Rhone, we saw new areas of cross-section and fission-process data declassified and released to the public domain. During this historic conference, I could not help but remember my earlier days as a University of Chicago freshman wondering just what that supersecret "Metallurgical Project" under the West Stands was all about. To me, the unprecedented open spirit of international cooperation that marked the first Geneva Conference was in stark contrast to the wartime secrecy that had of necessity characterized Fermi's historic first "criticality experiments" leading to achievement of the world's first self-sustaining fission chain reaction just thirteen years earlier in Chicago.

Two years after the first Geneva Conference in 1955, the International Atomic Energy Agency, a cornerstone of the "Atoms for Peace" implementation, was created (in October 1957) to focus on, and carry out, the promotion and control of the peaceful uses of nuclear energy in IAEA Member State countries around the world.

Fostered in large part by the Atoms for Peace program, throughout the 1960s peaceful nuclear energy programs flourished in many countries because supplier nations, including the United States, offered an extremely attractive long-term source of nuclear fuel, in part to discourage the development of other supply sources. Concurrently with this peaceful development, the 1960s also saw the number of nuclear weapons nations increase from three to five with the addition of France in 1960 and the People's Republic of China in 1964. These and other events led to steadily increased concerns about nuclear weapons proliferation --both the further build up within nuclear-weapons nations and especially the possible acquisition by new nations. In the mid-1960s, intensified efforts to reduce the risk of proliferation led ultimately, in 1970, to implementation of the Treaty on the Nonproliferation of Nuclear Weapons (NPT).

During this very active period of the mid-1960s, I had the unique (at that time) opportunity to serve for two years (1963-1965) with the headquarters staff of the International Atomic Energy Agency in Vienna, Austria. There I headed the Physics Section of the Division of Research and Laboratories, and our attention was initially focussed on the very active areas of fission and reactor physics; e.g., we organized and conducted the first in the ongoing series of IAEA Conferences on "The Physics and Chemistry of Fission" (Salzburg, 1965) as well as other IAEA Conferences on "Inelastic Scattering of Neutrons" (Bombay, India, 1964), and "Pulsed Neutron Research" (Karlsruhe, West Germany, 1965), among others.

During my assignment in Vienna, IAEA Director General Sigvard Eklund arranged for me

[a] To cite but one example in my specific area of delayed neutrons, such interactions provided an interesting and revealing historical perspective, namely that the key importance of delayed neutrons in controlling the rate of fission was recognized very early on, not only in the West, but also in the Soviet Union. The role of delayed neutrons in reactor kinetics and control was outlined in a very early prognosis of the prospects (peaceful and otherwise) of nuclear energy.[7] This remarkably forward-looking, if not "prophetic", paper was published in 1940--little more than a year after the discovery of fission, and more than two years before achievement of the first self-sustaining chain reaction. (Appropriately factored in, it could have brought more technical realism into some of the early estimates of how long it would take the USSR to develop the atomic bomb).

to meet with Werner Heisenberg who occasionally visited Vienna and "the Agency." In the course of our fascinating (for me at least) discussions --of physics and much more-- Heisenberg expressed interest and concerns in several areas, including the problem of effective "controls" over sensitive nuclear materials to ensure against their misuse (i.e., the general issue now called nuclear safeguards). A prospective future role for physics measurement instrumentation was implicit in our discussions, which left a lasting impression on me. Not only were my discussions with Heisenberg fascinating and inspiring on the technical level, but the same held true on the philosophical and personal level.[8] It seemed that Heisenberg (not unlike many thinkers) instinctively sought the larger connections between the various physical and biological sciences --and indeed between all the natural sciences and all other areas of human thought and inquiry, including what he called "the wider regions of life" as embraced by sociology, philosophy, and religion. In contemplating such a kind of epistemological unity, he points up the need for "breadth of thought," transcending narrow discipline boundaries and categories in such a way as to ultimately find a way back "to a natural balance between the spiritual and material conditions of life." Heisenberg was indeed a physicist of remarkable breadth and insight. After my return to the U.S. from Vienna I looked forward to future follow-on interactions with Prof. Heisenberg, particularly after I had become active in the application of physics measurement techniques to the burgeoning new field of nuclear safeguards. Sadly, however, this was not to be; Werner Heisenberg died on February 1, 1976. He has bequeathed to us all his monumental contributions to physics as well as his wonderful, and for me inspiring, essays and monographs[8] in areas of science, philosophy, and even a kind of noetic spirituality.

In July 1965, while still at the IAEA, I had the opportunity to tour and lecture in three major nuclear research centers in the USSR. One purpose of the IAEA-sponsored mission was to encourage and promote, through the framework of the IAEA, further international cooperation and exchange in certain unclassified areas of nuclear and reactor physics data. Extensive briefings and technical discussions were held with prominent nuclear and reactor physicists, including L. N. Usachev and S. P. Maksiutenko, the leading Soviet nuclear physicist in fission delayed neutron research (see Fig. 4). I found it both stimulating and reassuring that our Soviet counterparts shared many of our same concerns and problems in areas of reactor kinetics and control, as well as in criticality safety, and even the "coming" problem of safeguards and tight controls on nuclear materials.

Over the course of my two year assignment in Vienna[9], the impact of many factors --world nuclear developments, the new experience and perspective gained at IAEA headquarters, and my widening interest in international technical/political issues-- all seemed to combine to reaffirm more forcefully than ever before, my earlier convictions about the growing importance of achieving stringent safeguards and controls over sensitive nuclear materials, and the global challenge of nuclear nonproliferation generally. I also had felt for some time that the expanding capabilities of nuclear physics measurement techniques might well be brought to bear on this important problem. In retrospect, it seems abundantly clear that I myself had grown and evolved concurrently with the emerging issue and discipline of nuclear safeguards. In any case, by the time I returned to the United States in the Fall of 1965, I was firmly convinced that a vigorous R&D program should be

Figure 4. Fission and reactor physics discussion with Soviet physicists after the author's lectures at the Institute of Physics and Energetics in Obninsk, USSR. Left to right are V. Kuznetsov, head of the Department of Reactor Physics; the author; the late L. N. Usachev, distinguished theoretical reactor physicist; S. P. Maksiutenko, leading Soviet nuclear physicist in delayed neutron research, and physicist A. I. Abramov, USSR representative to IAEA International Nuclear Data Group.

[8] Just a couple of examples on the personal level: Heisenberg wore an absolutely unique tie-pin displaying a beautiful diamond-studded "h"--the celebrated mathematical symbol, called "h bar," known to every physicist the world over as the fundamental constant in the equation for the celebrated "Heisenberg uncertainty principle." When I exclaimed my admiration for the truly unique suitability of that particular tie-pin for him alone, Heisenberg beamed with eyes sparkling, "das ist ein geschenk von meinen studenten!" Among other delightful (for me) little sidelites, was the discovery, quite by chance, that we're fellow Sagittarians, both born on the German legendary "Krampus Day," December fifth.

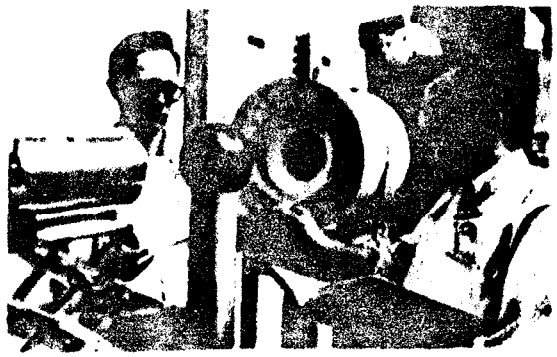

Figure 5. This 1967 photo from the early days of Safeguards R&D at Los Alamos shows Carl Henry and Chris Masters preparing to measure the delayed-neutron response of a uranium sphere using a "zipper" pulsed neutron generator (metal cylinder at left) as a source of interrogating neutrons.

launched to develop new techniques and instruments that would, in time, provide the technical basis for meeting the increasingly stringent safeguards requirements that some of us saw as "inevitable".

I felt strongly that the United States should take the lead in this key area, and that Los Alamos itself was actually quite unique in having both the expertise and the facilities that would be essential for the required R&D effort, including nuclear instrumentation and measurement know-how together with the full range of materials processing, fabrication, and recovery facilities for special nuclear materials. After some initial concerns about how an R&D program in an area such as safeguards (particularly international safeguards) would be regarded and supported at Los Alamos over the long haul, I decided to take the whole matter --my ideas, convictions, concerns, and all -- directly to the boss, Los Alamos Laboratory Director Norris Bradbury. (There was never any doubt, incidentally, that Bradbury --who directly succeeded Los Alamos' first Director, J. Robert Oppenheimer-- really was the boss, in the very best sense of the word).

After giving the matter due consideration, Dr. Bradbury came back with a very positive response, and proceeded to arrange for me to make presentations, briefings, etc. to Atomic Energy Commission Chairman Glenn Seaborg, Gerry Tape, and other AEC Commissioners, as well as to appropriate staffers of the Congressional Joint Committee on Atomic Energy, among others. In due course, funding was secured and the Los Alamos Safeguards R&D Program was launched on December 1, 1966. Six months later, the AEC established the Office of Safeguards and Materials Management at its Washington Headquarters, as well as a new Division of Safeguards in the AEC Regulatory Branch (now the Nuclear Regulatory Commission).

Figure 6. "THEN" and "NOW"--a span of nearly two decades. The upper photo, taken in October 1968, shows Los Alamos Laboratory Director Norris Bradbury and IAEA Director General Sigvard Eklund shortly after they had been introduced for the first time by Bob Keepin, Los Alamos Safeguards Group Leader. In the lower ("NOW") photo, taken during the recent 20th Anniversary Safeguards Symposium at Los Alamos, Bradbury, Eklund, and Keepin recall the early days of safeguards and Eklund's first visit to Los Alamos two decades earlier.

As already indicated, the selection of the Los Alamos Laboratory to spearhead United States leadership in safeguards R&D (both domestically and internationally) was due in large part to the unique facilities and expertise that Los Alamos already had in place. These included the full gamut of materials processing, fabrication and recovery facilities for special nuclear materials, including plutonium and uranium of all ^{235}U enrichments; it also had the world's leading (and original) expertise and remote-control facilities for experiments with, manipulation, handling and storage of fissionable materials of all compositions, shapes, and sizes in both subcritical and critical configurations.[a] In sum, the Los Alamos legacy of direct "hands-on" experience with fissionable materials (ranging from their detailed "microscopic" characteristics to their

[a] In connection with the special relationship between critical assemblies experiments and safeguards R&D, it is most appropriate, efficient, and mutually advantageous that these two professional disciplines are now closely coupled both technically and organizationally within the same technical division at Los Alamos (The Nuclear Technology and Engineering (N) Division).

overall "macroscopic" behavior in large critical systems, to the nitty-gritty of state-of-the-art processing technology) was anticipated to be uniquely valuable to the nation's pioneering safeguards R&D effort. And indeed, the internationally-recognized leadership of U.S. Safeguards R&D over the years bears out the highly productive synergism that has resulted from combining the special materials capabilities noted above with top-notch safeguards detection, measurement, and systems design expertise.

SAFEGUARDS TECHNOLOGY--PRESENT STATUS AND CAPABILITIES

Beginning in the 1960s, as nuclear activities expanded in many countries around the world, safeguards concerns increased correspondingly, and steadily greater requirements were placed on nuclear material measurement capabilities (e.g., with respect to detection sensitivity, timeliness, accuracy, and representative sampling) for the many forms and configurations of materials found in the nuclear fuel cycle. This, in turn, has led to the development and implementation of a new measurement technology to supplement, and complement, the traditional destructive assay methods of sampling and chemical assay. This new technology --now commonly known as nondestructive assay, "NDA"-- is based on direct physical measurements of unique characteristics of the fission process and of fissionable materials. As indicated earlier, there is a special synergism in the juxtaposition of nuclear fission and nuclear safeguards; i.e., the characteristic fission phenomena (such as gamma-ray and neutron emission) that are so basic to the fission process and the release of nuclear energy, provide at the same time their own unique "signatures" that are essential to effective measurement, accountability, and control of sensitive nuclear materials. Thus the development of modern safeguards technology has, by its very nature, entailed a synergistic combination of: (1) detailed knowledge of certain inherent characteristics of the fission process and fissionable materials together with (2) the practical application of these characteristics in the development of NDA instruments and techniques for safeguarding these materials.

Nondestructive assay techniques fall into two major categories, passive and active. Passive assay uses naturally emitted gamma-ray and/or neutron radiations as direct signatures of fissionable materials.[10] Active assay involves irradiation with neutrons or photons to induce fissions in the sample to be assayed. The resulting neutron or gamma-ray signatures are analyzed to determine quantitatively the amount of fissionable material present. For more than 20 years innovative safeguards research and development programs in the United States, and more recently in several other countries, have developed, tested, and implemented a broad range of passive and active NDA instruments and measurement/accountability systems that are now widely employed in safeguarding nuclear materials in nuclear facilities of all types. NDA instruments range in size and complexity from small portable units (e.g., as small as a carry-on brief case) for use by safeguards inspectors in on-site verification of nuclear material inventories, to large *in-situ* NDA measurement systems designed for routine in-plant use not only for safeguards and accountability, but also for process control, quality control, criticality safety, and radiological protection. In this section, we survey briefly the major categories of gamma-ray and neutron-based passive and active assay techniques, give representative examples of NDA instruments currently in use, and cite some notable instances of ongoing state-of-the-art NDA technique development.

First, in the area of passive gamma-ray assay, many different instruments have evolved employing the two well-known types of gamma-ray detectors; i.e., low resolution NaI(Tl) scintillation detectors and the high-resolution germanium solid-state detectors. Necessary corrections for sample attenuation are carried out using either an external gamma ray source or by suitable analysis of the measured response to the sample's own internal gamma rays. Gamma-ray measurements using the so-called "enrichment meter" principle are based on the fact that for fixed detector-sample geometry and for samples that are thick relative to the penetration depth of the 185.7-keV ^{235}U gamma rays, the count rate due to the 185.7-keV gamma rays is directly proportional to enrichment. When performed with care, NDA enrichment measurements can achieve 0.1 to 0.2% precision at one relative standard deviation.

In the case of plutonium isotopic composition measurements by gamma-ray spectroscopy, achievable accuracies are better than 1% for ^{241}Pu and ^{240}Pu, and better than 0.2% for ^{239}Pu. The well-known and widely used Portable Mini MCA, (Fig. 7) is a battery powered 2K/4K multichannel analyzer that can acquire, display, analyze, and record gamma-ray spectra from either NaI or high-resolution germanium detectors. Using suitable standards and calibration procedures the PMCA can provide accurate on-the-spot measurements of U enrichment as well as total ^{235}U content (and can also be used for some Pu verification applications). A second instrument, the Segmented Gamma Scanner[11], is used for measuring samples up to 200 liters in volume; it employs a transmission source that is viewed through a horizontal collimator slit to assay the sample as a series of horizontal segments, and then measures sample response and the transmission correction segment by segment. In

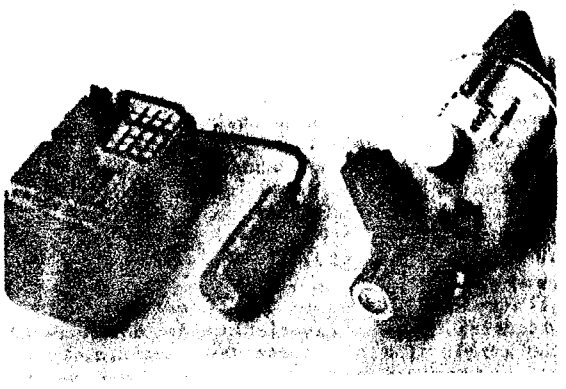

Figure 7. The Portable Mini-MCA (PMCA) is an "intelligent" battery-operated multi-channel analyzer (at left) that can display and record gamma-ray spectra obtained from a NaI detector (center) or a high resolution Germanium detector (at right). Using suitable standards and calibration procedures the PMCA can provide accurate on-the-spot measurement of U enrichment as well as total ^{235}U content, and can also be used for some Pu verification applications.

the case of solid materials (e.g. scrap and solid waste) an important source of bias can arise when lumps are present in the sample to be assayed; a method of detection and correction for the presence of lumps is under development that involves assaying the sample at different gamma ray energies.[11]

The second major category of NDA techniques is active gamma-ray assay, represented by the complementary techniques of gamma-ray densitometry and x-ray fluorescence. In the densitometer, a gamma-ray beam is passed through an assay sample and a gamma-ray detector measures the transmitted beam whose reduced intensity is a function of the gamma-ray energy and the amount, or concentration, of nuclear material between the source and detector. The isotopic sources, ^{57}Co and ^{75}Se-- with 122.0-keV and 121.1-keV gamma rays respectively-- nicely (and fortuitously) bracket the 121.7-keV K-absorption edge of plutonium. These sources are utilized in the so-called compact K-edge densitometer developed for in-line concentration measurements of Pu solutions in glove box lines without breaching or affecting in any way the glove box containment. An installed ^{57}Co-^{75}Se K-edge densitometer system has been used for nearly 10 years for assay of product solution in the analytical laboratory of the Tokai fuel reprocessing plant at Tokai-Mura, Japan.[12] Generally, the accuracy and precision of K-edge densitometer measurements are better than 1% and can approach 0.1%; in practice they are often supplemented with isotopic composition measurements. In the case of other elements (e.g., uranium, thorium) such fortuitous isotopic sources with gamma-ray energies that happen to lie just above and just below a desired absorption edge, generally do not exist, so x-ray generators are often used as the transmission source for densitometry measurements.

In the complementary technique of x-ray fluorescence (XRF), again a gamma-ray beam is passed through an assay sample, but here the absorbed, rather than the transmitted, gamma rays are used to provide an assay signal. The absorbing atoms are raised to excited states from which they decay by emission of x-rays; the energies of these x-rays are uniquely characteristic of the elements in the absorbing material, and their intensities are proportional to the amounts present. Gamma-ray densitometry and x-ray fluorescence have been applied most successfully to the measurement of uranium and plutonium concentrations in solutions. The two techniques are complementary; i.e., densitometry is best suited for SNM concentrations above ~10 g/l, whereas XRF is best suited for concentrations below this level. At least two hybrid assay systems have been built that combine densitometry and XRF. One is used to assay uranium and plutonium in light-water-reactor reprocessing solutions at Kernforschungzentrum Karlsruhe in the Federal Republic of Germany,[13] and the other is designed for routine use in the recovery section of the Los Alamos plutonium facility.[14]

Concerning advanced NDA technique development in the area of gamma-ray assay, two novel methods for determination of Pu concentration (and isotopic distribution) have recently been developed that require no external radioactive sources or x-ray generators, but rely only on the natural radiations from Pu. The methods are ideally suited to the assay of reasonably pure Pu solutions such as the product solutions of a reprocessing plant and the eluate solutions from anion exchange columns. The methods can be applied to aged or freshly separated Pu and can be used to measure Pu concentrations in pipes or tanks. The first method uses the MGA2 isotopic program developed at Lawrence Livermore National Laboratory.[15] In this program a relative detection efficiency curve is fitted from 59 keV to 208 keV including the discontinuity at the Pu K-absorption edge. For fixed sample thickness, the magnitude of the discontinuity is proportional to the Pu concentration of the solution. Applying this method to Pu solutions with concentrations ranging from 60 g/l to 320 g/l, it was found that the Pu concentrations can be determined to 1.9% with precisions of ~1.5%.

The second method[16] uses the ratio of a pair of gamma-ray or x-ray peaks from the Pu sample: one above the K absorption edge and one below the edge so that the absorption coefficients (mu) are substantially different. The mu values of 129 keV gamma (^{239}Pu) and 111-keV x ray (U K_β from ^{241}Pu) differ substantially, so the ratio of

these two lines is a strong function of Pu concentration, and for a fixed solution thickness the function can be used to determine Pu concentration from a measurement of the 111/129 ratio. Applying this ratio method to Pu solutions with concentrations ranging from 10 g/l to 320 g/l, Pu concentrations were determined to 0.26% with precisions of ~0.2%. Calculations show that while the ratio method is insensitive to the amount of low Z absorber (Z < 10), for best results the medium Z matrix (Z < 40) in the solution should be less than 6% of the Pu concentration, and the high Z matrix should be less than 3% of the Pu concentration. Thus if the concentration of impurities in the Pu solution is less than the amounts given above, the method can be used to determine Pu concentrations from 10 gm/l to 300 gm/l with less than 1/2% bias. When the solution is very thick, the ratio approaches a unique asymptotic value, with the very practical consequence that the ratio method can therefore be used to determine Pu concentrations in tanks or bottles without drawing samples.

Turning now to neutron-based NDA techniques, we address first passive neutron methods, an area where once again we find close technical coupling between basic characteristics of the fission process and the development of state-of-the-art NDA instrumentation. Neutrons originating in nuclear materials are primarily due to (1) spontaneous fission (largely in Pu-238, 240, and 242) and (2) (α,n) reactions in light elements (e.g., in the commonly used compounds of uranium and plutonium, notably the oxides, carbides, and fluorides, or in B, Be, or Li impurities). An additional source of neutrons can arise, especially in larger samples, from induced-fission multiplication in the sample. In general, passive neutron detection provides a convenient assay measurement, especially for plutonium samples, because of high neutron yields, detector simplicity, and neutron penetrability through the sample and storage or shipping containers. The most frequently used neutron detector for NDA instrumentation is the ^{3}He proportional counter, chosen for relatively high neutron detection efficiency, insensitivity to gamma rays, reliability and long-term stability. Spontaneous fission "coincident" neutrons are distinguished from (α,n) "singles" neutrons by coincidence counting techniques based on high resolution (ultra fast) "shift register" coincidence electronics.[17]

The two most frequently used passive neutron signatures are the spontaneous-fission decay of the even-numbered plutonium isotopes and the (α,n) reaction in UF_6. Neutron coincidence counters measure the total spontaneous-fission decay rate from plutonium, which includes contributions from the three even-numbered isotopes ^{238}Pu, ^{240}Pu, and ^{242}Pu. Generally, ^{240}Pu is the major contributor, and it is convenient to define the quantity:

$$^{240}Pu(eff) = 2.52\ ^{238}Pu + {}^{240}Pu + 1.68\ ^{242}Pu,$$

where the coefficients 2.52 and 1.68 are determined from fundamental measurements of fission neutron emission from various fissionable isotopes, and take account of the higher spontaneous fission decay rates as well as the higher average number of neutrons per fission ($\bar{\nu}$) in ^{238}Pu and ^{242}Pu. Total plutonium can then be calculated from plutonium isotopic composition; e.g., as determined from high-resolution gamma-ray spectroscopy. As extensive field experience has demonstrated, the combination of these two techniques can be extremely effective --as long as the contribution from (α,n) neutrons and sample self-multiplication is not too large (cf discussion of (α,n) and multiplication effects below).

For most applications the passive neutron signal from uranium is too small to provide a reliable assay signature. The major exception is UF_6 where the high cross section of the (α,n) reaction on fluorine provides a useful uranium assay signature, that has been used to measure highly enriched UF_6 cylinders and liquid UF_6 at the product load-out point of enrichment plants.[18]

By far the most widely used passive neutron counter is the High Level Neutron Coincidence Counter, HLNC[19] (See Fig. 8) developed at Los Alamos for the assay of bulk plutonium samples ranging from 10 g to several kilograms of plutonium, and ^{240}Pu content from a few per cent to ~ 30%. The HLNC has become one of the real "workhorses" of IAEA safeguards inspection and verification operations worldwide. It can assay samples containing 500 g or more of plutonium in 300 seconds with a precision and accuracy of better than 1%. The utility of the basic HLNC system has been greatly extended by the development of a whole family of HLNC-like detectors with specialized detector heads, but all employing the same basic "shift-register" coincidence electronics.[17] Individual detector heads vary greatly depending on the materials and configurations to be measured (e.g., ranging from heads for small inventory samples to large fast reactor fuel assemblies).

Many nuclear material samples exhibit a measurable neutron multiplication value, especially the larger samples with hundreds or thousands of grams of either ^{235}U or ^{239}Pu. Thus passive neutron measurements can be altered by neutron moderators (e.g., moisture), reflectors, and absorbers in or near the sample. Conventional coincidence counting procedures have worked

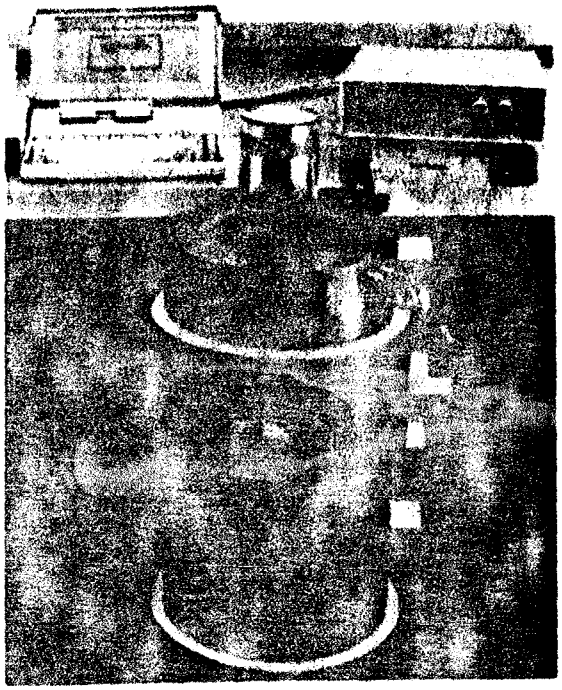

Figure 8. The High-Level Neutron Coincidence Counter, widely used for assay of plutonium samples ranging from 10 g to several kilograms with measurement precision and accuracy of ~1% or better. The HLNC is used routinely by IAEA and EURATOM safeguards inspectors in nuclear installations around the world.

reasonably well for pure PuO_2 materials; however for highly multiplying samples, impure oxides, samples with high ^{241}Am content, and salts with high (α,n) yields, the procedure fails because of the unknown multiplication and induced fission rates. A method was developed several years ago to correct for multiplication effects based on measurement of the real coincidence count rate, R, together with the ratio of R to total neutron rate, T; i.e., the "reals to totals" ratio.[20] More recently, detailed multiplication corrections have been applied to special cases; e.g., a neutron self-interrogation technique for assay of plutonium in high (α,n) materials.[21] Also, Monte Carlo simulations of neutron coincidence counter assays have been successfully applied[22] to passive assay of large, moist PuO_2 samples for which erroneously high assay values are obtained by conventional coincidence counting procedures.

Notwithstanding the significant progress that has been made in developing coincidence counting corrections, the major area of development in neutron assay techniques continues to be focused on finding better ways to measure and correct for sample multiplication effects. To state the problem briefly: in the general case of passive neutron counting there are three principal unknown variables: plutonium mass, sample self multiplication, and (α,n) rate; however, there are only two measured parameters in conventional coincidence counting i.e., "real" neutron coincidence count rate, R, (coincidence neutrons that originated in a common fission event) and total neutron count rate, T. Among a number of possible approaches,[23] this basic problem of "one-too-many unknowns" is currently being addressed at Los Alamos in two quite different ways, although each involves the development of an innovative neutron counting system. One is a fast neutron counter using liquid scintillator detectors, and gamma ray/neutron pulse shape discrimination.[24] This detection system is designed to measure all three of the unknown quantities noted above, and to minimize any interference from gamma-ray response of the scintillators. High-resolution coincidence circuitry separates the amplified scintillator pulses into single, double, and triple coincidence events, just as neutrons are emitted in pairs and higher multiplicities in the fission process itself.

The second innovative neutron counting system is the "neutron multiplicity counter"[25] designed to investigate the use of neutron multiplicity distributions for NDA of plutonium samples. Like other NDA methods, this new approach to neutron assay of plutonium is also based on an inherent characteristic of fission physics --namely that the average number of prompt neutrons produced in the ^{240}Pu fission process is higher for the neutron-induced fission of ^{239}Pu than for the spontaneous fission of ^{240}Pu. Based on this small but telling characteristic difference, the detailed measurement and analysis of neutron multiplicity distributions can be used to determine the neutron multiplication in plutonium samples. To enhance the multiplicity conformation, the multiplicity counter was designed with low deadtime, fast neutron die-away time, and high efficiency. The working system (overall dimensions ~80-cm diam by ~70-cm high) is shown in Fig. 9. The neutrons are detected by a total of 130 ^3He detectors configured in five concentric rings; the sample cavity is 15-cm in diameter, and the body of the counter is surrounded by 5 cm of polyethylene reflector. A total of 34 AMPTEK circuits[26] are used to reduce deadtime. Neutron multiplication has been determined by analysis of measured neutron multiplicity distributions for samples of pure plutonium oxide with varying ^{240}Pu fractions (5-20%), for plutonium oxide mixed with matrix materials having various (α,n) yields, and for pure and impure plutonium metal. Mass uncertainties from counting statistics range from 0.3% for ~1 Kg pure Pu metal to 1.4% for ~1 Kg pure PuO_2 with (α,n)/SF neutron ratios below ~1.

Figure 9. The "neutron multiplicity counter" in operation at the Los Alamos safeguards R&D laboratory. The outer shield measures ~80 cm diameter by ~70 cm in height and the central sample cavity is 15 cm in diameter. The neutron multiplicity counter is designed for accurate nondestructive assay of Pu samples with unknown self-multiplication and (α,n) yields.

(SF=Spontaneous Fission). Counting statistics errors rise rapidly for high (α,n) samples ((α,n)/SF > 3), and at high counting rates. The most promising applications of the neutron multiplicity counter are assay of impure metal samples up to several Kg and impure oxide samples up to ~1 Kg, both with (α,n)/SF ratios < 2. Concerning possible complementarity between the liquid scintillator and neutron multiplicity methods, for high (α,n) materials the liquid scintillator counter, when fully developed, may provide a useful supplement to the demonstrated capabilities of the neutron multiplicity counter.

Moving now from passive to active neutron assay, here the fission process itself is employed directly to stimulate (or "induce") a desired assay signature. The NDA of ^{235}U materials provides a very practical case in point, because ^{235}U does not have a passive neutron signature, ^{235}U-bearing samples are irradiated with neutrons to induce fissions in the ^{235}U, and the resulting emitted fission neutrons (prompt and/or delayed) provide a signature for accurate NDA. Examples are given below of state-of-the-art active neutron NDA instruments that utilize prompt neutron as well as delayed neutron response measurements (and also delayed gamma-ray response) to determine fissile material content with attainable accuracies of one percent or better.

Figure 10. Indicative of ongoing U.S. technical support to the International Atomic Energy Agency, this front cover of the IAEA Bulletin shows a top view of the universal fast-breeder-reactor assembly counter developed at Los Alamos to measure total Pu content by neutron coincidence counting. The advanced analog electronics can accommodate neutron counting rates above a million counts/ second, thereby enabling accurate measurement and verification of Pu in fast-breeder-reactor assemblies containing as much as 16 Kg of reactor grade plutonium (20% ^{240}Pu).

First we cite the Active Well Coincidence Counter (AWCC) used for assay of ^{235}U content in enriched uranium materials. Two (α,n) neutron sources (AmLi, each ~5×10^6 n/s) located above and below the sample well are used to interrogate the sample, and the induced fission neutrons are counted with standard shift register coincidence electronics. Coincidence counting discriminates against the random "singles" (α,n) neutrons from the AmLi sources while detecting coincident neutrons from neutron-induced fissions in the ^{235}U present in the sample. The AWCC is used to measure bulk UO_2 samples, high enrichment uranium metals, LWR fuel, pellets, ^{233}U-Th fuel materials having high gamma-ray backgrounds, and more recently even mixed-oxide samples.[27] A second important application of active neutron coincidence counting is the Uranium Neutron Coincidence Collar (UNCL, see Fig. 11). The UNCL can be operated in both the active and the passive mode to measure ^{235}U and the ^{238}U content, respectively, of both PWR and BWR light-water reactor assemblies. In the active mode a low-intensity (5 x 10^6 n/s) AmLi neutron source interrogates the fuel

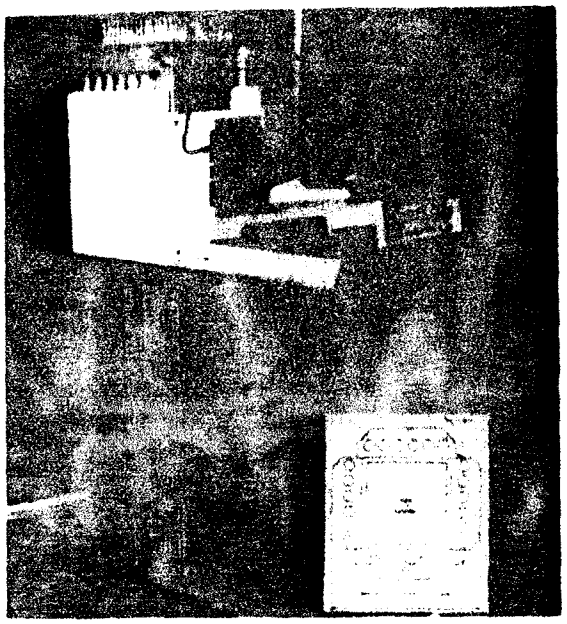

Figure 11. The Uranium Neutron Coincidence Collar (UNCL) is shown here measuring a PWR fuel assembly mockup. The UNCL can be operated in both the active and the passive mode to measure ^{235}U and ^{238}U content, respectively, of both PWR and BWR light-water reactor assemblies. The ^{235}U response sensitivity enables detection of the removal or substitution of 3-4 rods in a PWR assembly and one rod in a BWR assembly.

assembly, and the induced prompt neutrons (from ^{235}U fission) are coincidence counted. When no interrogation source is present, the passive neutron coincidence rate (from ^{238}U spontaneous fission) gives a measure of the ^{238}U in the fuel. The ^{235}U response sensitivity enables detection of the removal or substitution of 3-4 rods in a PWR assembly and one rod in a BWR assembly.

The so-called ^{252}Cf shuffler[28] illustrates the application of active neutron interrogation together with delayed neutron response measurements. The heart of the ^{252}Cf shuffler is an annular neutron detector into which the sample to be assayed is placed. A large ^{252}Cf source (10^{7} to 10^{10} n/s) is repetitively cycled ("shuffled") into and out of the detector cavity region to irradiate the sample and induce fissions in the ^{235}U present. Between successive ^{252}Cf neutron irradiations the detector is gated "on" to count delayed neutrons from the induced ^{235}U fissions. Properly calibrated, this delayed neutron signal then provides a measure of the amount of ^{235}U in the sample. The shuffler technique has been adapted to different measurement problems and container sizes from small vials to 200-ℓ (55-gallon) drums. Shuffler systems can measure highly-radioactive samples, such as irradiated fuel and reprocessing waste, because the ^{252}Cf source strength can be increased somewhat to override the background radiation. Fig. 12 illustrates a ^{252}Cf shuffler system installed at the UK prototype fast reactor reprocessing plant in Dounreay Scotland under a joint evaluation project between the U.S. and the U.K. The shuffler system has been used nearly continuously over the past five years for the assay of plutonium in hot scrap and leached hulls (from spent fast reactor fuel) in the head end of the reprocessing plant. In several applications in the U.S. as well as in Dounreay, the shuffler has been built into an existing hot cell, and the shielded source "storage position" is located in the center of the existing cell walls. Another large ^{252}Cf shuffler system has been installed at the Fluorinel and Fuel Storage ("FAST") Facility in Idaho, where it is in routine use for fissile assay of irradiated, highly enriched uranium fuel assemblies (with assay precisions of 2-3%).[29]

The ^{252}Cf shuffler systems just noted provide yet another practical example of the common technology "roots" of nuclear fission and nuclear safeguards; i.e., the unique characteristics of fission delayed neutrons (decay group periods, abundances, and absolute fission yields) that pace the fission chain reaction and enable the precision control of nuclear reactor kinetics are the very same unique delayed neutron characteristics that provide the incisive "signatures" required for

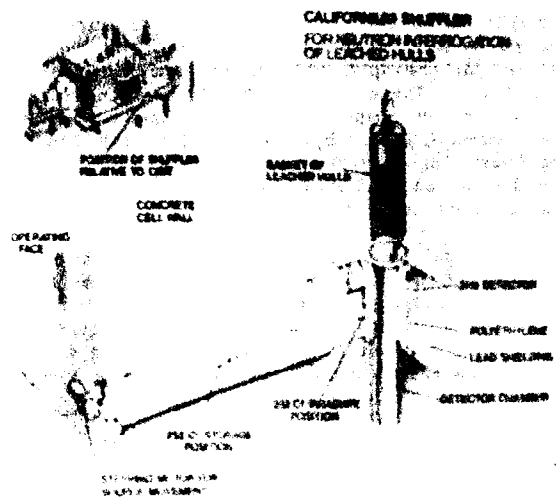

Figure 12. The ^{252}Cf Shuffler used for neutron interrogation and delayed neutron assay of plutonium in leached hulls (from spent fast reactor fuel) in the head end of a reprocessing plant. The Shuffler can also be used for routine precision assay of ^{235}U content in irradiated highly-enriched uranium fuel assemblies.

accurate delayed neutron assay of fissile materials for safeguards.

Finally we cite an active neutron interrogation system that utilizes not delayed neutron but delayed gamma-ray response: the automated ^{252}Cf fuel rod scanner, developed early in the Los Alamos Safeguards R&D program, is used for quantitative assay of both light-water-reactor and fast-breeder-reactor fuel rods. The fuel rods are irradiated with a ^{252}Cf neutron source to induce fission in the fissile fuel (^{235}U or ^{239}Pu) loading of the rods. Measurements on the delayed gamma rays from induced fissions in the fuel rods are then used to determine pellet-to-pellet uniformity of loading, and total fissile content; i.e., grams ^{235}U or ^{239}Pu, to better than 0.5% accuracy. Fuel rod scanners are today widely used --for process and quality control, as well as material accounting and control-- in commercial nuclear fuel manufacturing plants in many countries.

Many of the instruments described in this paper exemplify an important trend in NDA instrumentation development, namely computerization and standardization of measurement equipment and procedures for safeguards inspection and verification. Insofar as possible the new, "intelligent" NDA instruments are equipped with software programs for performance self-diagnostics, calibration and measurement quality control. Some instruments, such as the Portable Mini-MCA, also feature interactive-display prompting of the user (e.g., safeguards inspector) through the proper detailed measurement procedure, and perform all necessary calculations to give direct on-the-spot measurement and verification results. These "intelligent" NDA instruments offer many important advantages in field performance (e.g. by IAEA inspectors), in new equipment acceptance and inspector training, as well as significantly reduced equipment maintenance and field-logistics problems.

Much of the current NDA development effort is directed toward modifying and improving existing techniques; e.g., improved methods for neutron multiplication correction, gamma-ray peak area evaluation, and gamma-ray attenuation in heterogeneous materials, as well as ongoing development and improvements in the important NDA area of calorimetric assay techniques and procedures.[10] One example of work on new technique development is the application of laser-induced breakdown spectroscopy to high-sensitivity measurements of flowing uranium and plutonium solutions as well as to highly-radioactive solutions.[30] Clearly a key area of ongoing concern in safeguards R&D is the development and field implementation of good measurement standards and calibration procedures, and the accurate determination of bias and precision for NDA techniques. Noteworthy in this connection is the use of Monte Carlo simulations[31] to determine calibration parameters for neutron coincidence assay of mixed oxide fuel elements, and the potential for more effective, less costly inspector verification of finished reactor fuel elements by reducing reliance on expensive physical standards.

In addition to transportable and in-plant NDA systems for quantitative measurement of SNM, there also is an active ongoing effort in the development of rugged, hand-held instruments for use by relatively untrained personnel for search and detection of special nuclear materials. For instance, two recently developed instruments provide the capability for direct, on-the-spot verification of the presence or absence of certain sensitive nuclear materials.[32] One instrument uses a $^{6}LiI(Eu)$ scintillator and pulse-height analysis to verify the presence or absence of plutonium by measuring neutrons emanating from a container surface. The other instrument uses an LED-stabilized NaI(Tl) scintillator and three single-channel analyzers to measure and strip Compton background from a gamma-ray peak or region of interest to verify that certain isotopes of plutonium or particular enrichments of uranium are present or absent. These new instruments are lightweight, have low power requirements, and are easily operated in the field by nonspecialists.

The nation's Safeguards R&D program is committed to the development and application of state-of-the-art NDA instruments, techniques, and systems to meet the requirements of government and commercial nuclear facilities, as well as the needs of safeguards inspection authorities, both domestic and international. A highly productive cooperative R&D effort between instrument developers, safeguards systems analysts, and materials processing experts is actively ongoing today with the overall objective of developing integrated "near-real-time" material accounting and control systems for demonstration, test and evaluation in various facility types. A timely case in point is the recently installed integrated system of automated NDA instrumentation (gamma-ray spectrometers supplied by Livermore, neutron coincidence counter by Los Alamos, and calorimeters by Mound Laboratories) for nuclear materials accounting and process control in the new plutonium scrap recovery facility at the U.S. Department of Energy's Savannah River Plant in South Carolina (See Fig. 1)). The entire integrated NDA system is presently undergoing full-scale test and evaluation at the Savannah River recovery facility.

Finally, we cite one further example of ongoing safeguards technical support and cooperative activities at the international

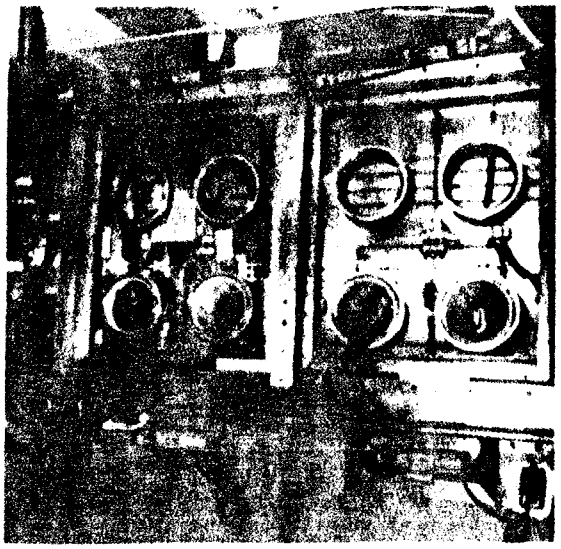

Figure 13. High resolution gamma ray spectrometers (for solids isotopic analyses) recently installed in the sample assay room of the new plutonium scrap recovery facility at the DOE Savannah River Plant. (Upper and lower photos are front and rear views of glove box line). The pictured spectrometers are part of a complete integrated NDA system, which also includes a feed assay neutron coincidence counter and four alarmeters.

level. Under a formal US-Japan agreement for cooperation in the peaceful uses of nuclear energy, a number of NDA instruments are currently being developed by the US Safeguards R&D program in cooperation with the Japanese PNC (Power and Nuclear Fuel Development Corporation), these instruments will be used for in-line measurement of mixed oxide (MOX) fuel in the new, large (5 ton MOX/year output) Plutonium Fuel Production Facility (PFPF) at Tokai Mura, Japan.[33] The PFPF facility will supply MOX fuel for Japan's fast breeder reactors, MONJU and JOYO, as well as future plutonium-recycle light water reactors. NDA instruments to be installed in the PFPF will measure feed materials, process materials, fuel pellet fabrication, handling and transfer, fuel pins in trays, completed MOX fuel assemblies, as well as process-line holdup, scrap and waste. All material handling and processing operations are carried out by automated, remote control so that all the in-process MOX material is, in effect, confined within a sealed "containment envelope" from the input of feed material to the final output of finished MOX fuel assemblies. The PFPF facility represents a very significant advancement in modern nuclear fuel fabrication technology and, as such, represents a correspondingly significant challenge and opportunity for the development, test, and implementation of state-of-the-art safeguards technology in a state-of-the-art high-throughput nuclear production facility.

Notwithstanding the impressive progress that has been made in safeguards technology development and implementation, it is patently clear that the effectiveness of nuclear safeguards depends not only on technology and hardware, but also on the people involved --both the safeguards inspectors and the "inspectees" in nuclear facilities. As in all human endeavors, the actual implementation of effective and workable safeguards must be carried out by *people* --and moreover by qualified people with the requisite training, knowledge, and motivation. Toward this absolutely essential goal of effective safeguards training and technology transfer, the United States has led the way in developing and conducting over a dozen safeguards training courses each year for inspectors and safeguards professionals from throughout the United States and countries around the world. Indicative of the importance attached to safeguards training and technology transfer, since 1980 every new IAEA (International Atomic Energy Agency) inspector has been required to complete the Los

Alamos NDA training course for IAEA inspectors. To date this has involved a total of some 400 IAEA people.

Effective operation of the overall international safeguards regime depends not only on a well trained IAEA inspectorate, but also on the effectiveness of the State (i.e., national) safeguards systems whose performance the international system must independently verify. It is therefore essential to have in place an ongoing program of training and technology transfer for key personnel in Member States who are responsible for the State's safeguards system (including, of course, safeguards at the State's nuclear facilities), and for the interface between the State system and the IAEA. The need for steadily improved State Systems of Accounting for and Control of Nuclear Material ("SSAC") led to the series of IAEA Basic SSAC Training Courses that were begun by the Agency in 1976; in recent years these have been strongly augmented by SSAC implementation courses given alternately in the U.S. and in the USSR, both in close collaboration with the IAEA. The 1988 SSAC course (focusing on discrete item facilities) was convened November 14-26, 1988, at Tashkent, Uzbekistan, in the USSR. It is followed in the USA by an advanced SSAC course (focusing on bulk handling facilities) held in Los Alamos/Santa Fe, New Mexico and Richland, Washington, from May 1-19, 1989 (See Fig. 14). Course participants include a total of 24 course attendees (trainees) from 20 nations, and 32 lecturers from 7 nations, the IAEA in Vienna, and the EURATOM Safeguards Directorate in Luxembourg. Many years of experience have shown that the SSAC and IAEA inspector courses as well as other international and domestic safeguards training courses contribute not only to the technical effectiveness, acceptance, and credibility of safeguards, but also help to build a spirit of cooperation, mutual confidence, and a shared sense of professional commitment among safeguards professionals from around the world. By its very nature and mission, the safeguards profession tends to engender a common dedication to the challenge of nuclear safeguards and nonproliferation in the nuclear age whose 50th anniversary is commemorated in this historic international conference.

Concerning the nuclear age and prospects for the future, I would offer a general observation that the IAEA's unique world-wide safeguards inspection and verification experience can provide a useful guideline for development and evaluation of possible future international verification systems, as for example in the sensitive and pivotal area of nuclear arms control. As IAEA Director General Hans Blix has pointed out in reference to IAEA safeguards, the success of this first bold experiment in institutionalized international verification could serve as an inspiration and valuable guideline for developing needed verification and control measures in the context of nuclear disarmament--or conversely, should international safeguards be perceived to falter, this could be a significant setback in the prospects for nuclear arms control. With US-USSR

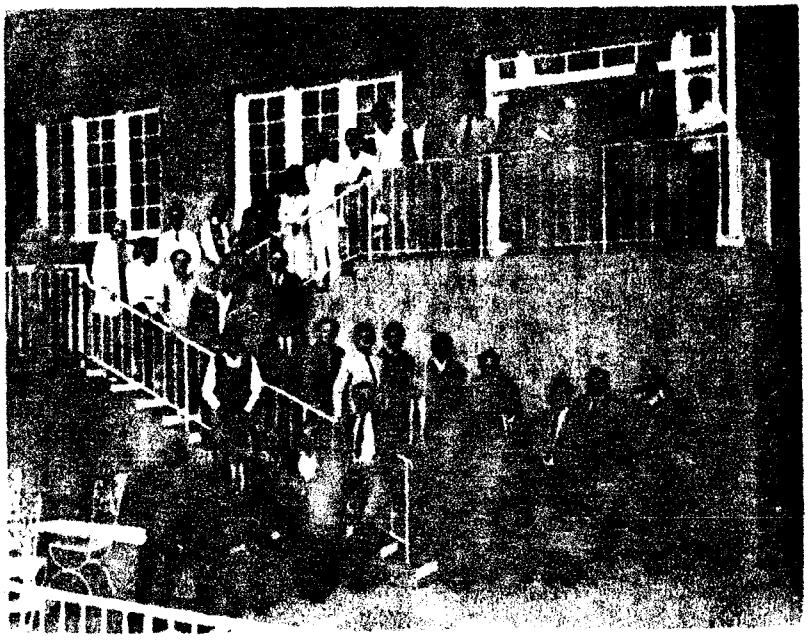

Figure 14. The most recent in the ongoing series of International Training Courses on State Systems of Accounting For and Control of Nuclear Materials was held May 1-19, 1989 in the USA at Los Alamos/Santa Fe., N.M. and Richland, Wash. Course participants, shown in this group photograph, include 24 course trainees from 20 nations, and 32 lecturers from 7 nations, the IAEA, and the EURATOM Safeguards Directorate in Luxembourg.

implementation of the INF treaty already well underway, and reasonable prospects for further effective arms control negotiations, it seems abundantly clear that modern safeguards technology, suitably adapted as necessary, is destined to play a key role in the achievement of effective verification of nuclear arms control agreements, of whatever type the future may bring.

The preparation of this review naturally involved extensive reflection on the history of nuclear fission and the consequent emergence of the issue and the discipline of nuclear safeguards; in the course of all this reminiscing I could not help but recall, with nostalgia and gratitude, the many outstanding friends and associates who provided inspiration, guidance, and support at various stages along the way. Although it would be impossible to name all those to whom I am thus indebted over the years, this unique 50th Anniversary Conference affords a very special opportunity to express my debt of gratitude to both of our distinguished Conference Co-chairmen: to *Professor Emilio Segre*, my postdoctoral advisor and sponsor at Berkeley in the early 1950s, and to Professor Glenn Seaborg who, as AEC Chairman, helped ensure establishment of the nuclear safeguards R&D program at Los Alamos over two decades ago. It has been a great privilege for me as for many of us in the nuclear field, to be associated with both of these great scientist-pioneers of nuclear fission.

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