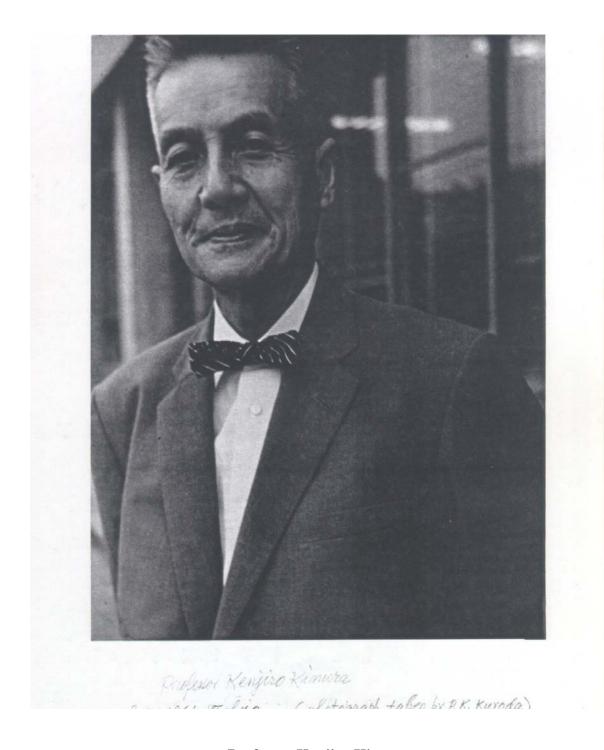
My Early Days at the Imperial University of Tokyo

Paul Kazuo Kuroda

[This manuscript was transcribed at the University of Missouri-Rolla from a manuscript that Professor P. K. Kuroda mailed to Dr. Ramachandran Ganapathy on 10 January 1992. Mrs. Louise Kuroda, Ramachandran Ganapathy, and Oliver Manuel proof-read the manuscript. Oliver Manuel wrote the Figure captions and the Appendix. He is responsible for any typographical errors that remain.]



Professor Kenjiro Kimura

Paul K. Kuroda took this picture of Professor Kenjiro Kimura in June 1961 in front of the Tokyo office of the Institute of Atomic Energy Research.

CHAPTER 1 HISTORICAL INTRODUCTION

The Eleventh Year of Showa: 1936

Shortly after 1:00 p.m. on Monday, April 13, 1936, about twenty newly admitted students at the Department of Chemistry of the Imperial University of Tokyo stood in attention and bowed to a young professor who walked into the room to deliver his opening lecture in analytical chemistry. The name of the professor was Kenjiro Kimura. He was born on May 14 of the year 1896, two months after Henri Becquerel (1852-1908) in Paris discovered radioactivity. He was only 39 years old and the youngest of all the full professors of the Chemistry Department.

Monday afternoon at 1:00 p.m. was not a regularly scheduled time and besides, it was the time of cherry blossoms in Tokyo. We had anticipated that perhaps the first lecture would be brief. We were in for a big surprise however, because Professor Kimura went on for a full two hours, writing on the blackboard the titles and the names of the authors of no less than 40 reference books, briefly commenting on the features of each book (See Fig. 1). Twenty-one of these books were written in English, twelve in German, four in French, and the remaining four in Japanese, including one which was written by Professor Kimura himself. While taking notes of the names of all these reference books, I felt that I could not possibly read all these books, especially those written in French, a language I had never studied before. Fortunately however, the reference book most highly recommended by Professor Kimura was "Applied Inorganic Analysis" by W. F. Hillebrand and G. E. F. Lundell. When I began my teaching and research career at the University of Arkansas many years later, I purchased the second edition of "Applied Inorganic Analysis" by Hillebrand, Lundell, Bright and Hoffman of the U.S. National Bureau of Standards, published by John Wiley and Sons, Inc., New York in 1953. I found this book to be extremely useful, especially for radiochemists working on new

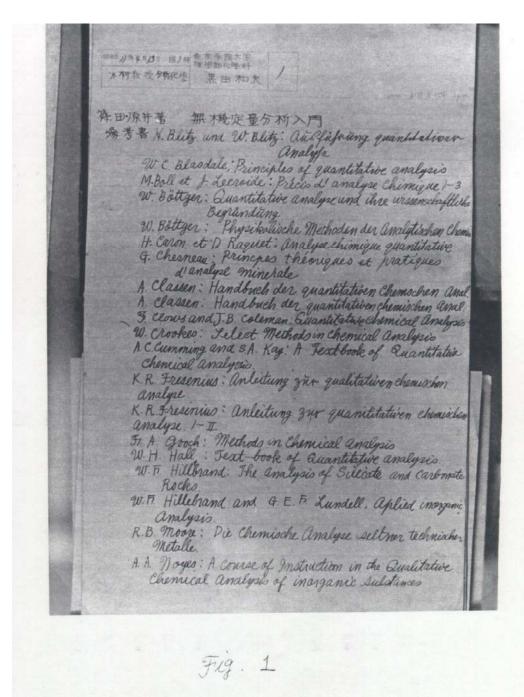


Figure 1. This is a copy of the list of reference books Professor Kimura assigned to his analytical chemistry class in their first meeting on 13 April 1936. P. K. Kuroda kept this list as part of the Kimura lecture notes, *Lehrbuch der Analytischen Chemie*, that he had bound as a book. See Fig. 5.

procedures for isolating and determining the amounts of radionuclides in minerals, rocks, ores and natural waters.

The next lecture, which was held at the regularly scheduled time, from 10:00 a.m. to noon, on Wednesday April 22, 1936, was devoted to the history of analytical chemistry. The lecture, which lasted a full two hours, covered a period of 300 years, starting with the work of Robert Boyle (1627-1691), who used the word "analysis" for the first time, up to the developments of new branches of analytical chemistry in the 20th century, for example the concept of "radioactive indicator", established by George de Hevesy, under whom Professor Kimura studied in Copenhagen, Denmark, in the 1920's.

The third lecture was held from 10:00 a.m. to noon on Wednesday May 6, 1936, because Wednesday April 29 was Emperor Hirohito's Birthday, which was one of the most important national holidays of Japan. On this day, Professor Kimura began a series of lectures on qualitative chemical analysis, which covered, aside from the classical systematic method of qualitative analysis, topics such as microscopic or microchemical qualitative analysis and spot tests, spectrochemical analyses, flame spectroscopy, emission spectra, luminescence, and absorption spectra (May 13 through June 3). (See Fig. 2).

On Wednesday June 10, 1936, he introduced us into the field of x-ray spectroscopy, which was the area in which he was generally regarded as the top leader in Japan.

One day at about this time, a notice was placed on the bulletin board at the main entrance of the Chemistry Building. It was the announcement that Dr. Francis William Aston from Cambridge University in England will be visiting our university and giving a special lecture on "Mass-Spectra and Isotopes" at 4:00 p.m. on Saturday June 13, 1936. I knew that Dr. Aston was the recipient of the 1922 Nobel Prize in Chemistry "for his discovery, by means of his mass-spectrograph, of isotopes in a large number of non-radioactive elements, and for his enunciation of the whole-number rule". He invented his

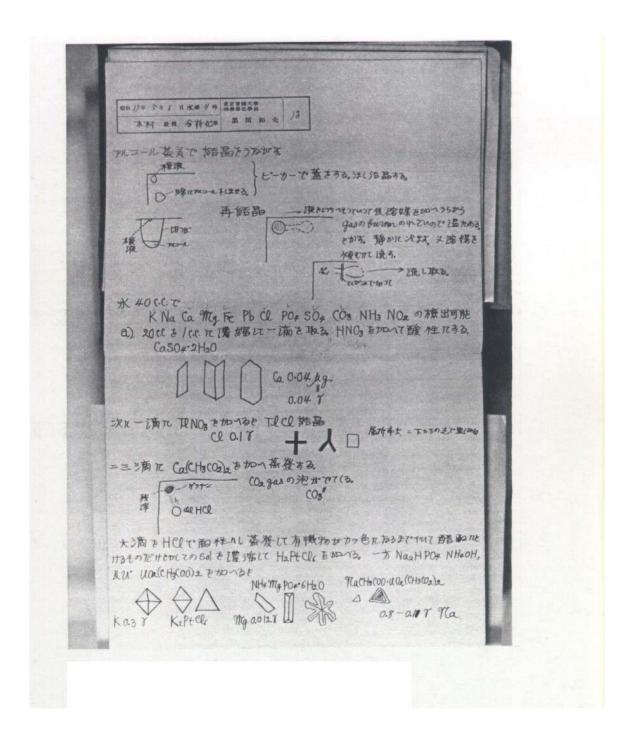


Figure 2. This is a copy of Kuroda's notes from Professor Kimura's lecture on qualitative chemical analysis in the spring of 1936.

mass-spectrograph as early as in 1919 and yet, for some reason, there was not a single mass-spectrograph in operation at the Chemistry and Physics buildings of the Imperial University of Tokyo in 1936.

I felt that the mass-spectrograph would become an extremely important tool for analytical chemistry of the future. With this new apparatus, it might become possible to measure not only the total amounts of various elements in the sample, but also the relative abundances of all the stable isotopes of the elements. I remembered seeing "Mass-Spectra and Isotopes" by F. W. Aston in the Maruzen Book Store in the Nihonbashi district of downtown Tokyo and the price of the book was listed as 12 Yen and 60 Sen, which corresponded to about one-fourth of the monthly starting salary of 55 Yen paid to a junior assistant at the University in those days. I decided however, to purchase this book before the forthcoming lecture of Dr. Aston and managed to obtain the money from my father.

In the afternoon of Friday June 12, 1936, I went to the Maruzen bookstore, purchased the book and began reading it. I could not finish reading the 248-page book before the lecture began at 4:00 p.m. in the afternoon of Saturday, June 13th. I am reasonably certain, however, that I went as far as Chapter VIII, Method of Precision, where on page 82 Aston explains the definition of "packing fraction", because when the lecture ended, the first question someone from the audience asked Dr. Aston was "What is meant by packing fraction?" The person who asked the question appeared to be a physicist in his 30's and I felt sorry for him, but also for Dr. Aston, who had to again explain the meaning of this important concept.

Among the many things I learned from the visit of Dr. Aston in 1936, what interested me the most was the idea which he expressed in the final paragraph of his Nobel Lecture entitled "Mass-Spectra and Isotopes" on December 12, 1922. He stated: "Should the research worker of the future discover some means of releasing this energy in a form which could be employed, the human race will have at its command powers beyond the dreams of scientific fiction; but the remote possibility must always be

considered that the energy once liberated will be completely uncontrollable and by its intense violence detonate all neighbouring substances. In this event the whole of the hydrogen on the earth might be transformed at once and the success of the experiment published at large to the universe as a new star". Obviously, he was anticipating the potential use of atomic energy for peaceful purposes, but he was also fearful of its use as a nuclear weapon which might lead to the destruction of the world.

I never dreamed at that time that the atom bombs built by American scientists would be used against the people living in Hiroshima and Nagasaki only nine years later, in the summer of 1945. The fear expressed by Aston that the nuclear energy once released may be uncontrollable and involve the whole of the hydrogen on the earth appeared to me fascinating, although most of the scientists living in Japan at that time seem to have dismissed the idea as a mere fantasy. It is interesting to note here, that I published the theory of the Pre-Fermi natural reactor twenty years later in the *Journal of Chemical Physics*, volume **25**, page 781 (1956) and that the idea behind this theory had its origin in my encounter with Dr. Aston in Tokyo in 1936.

During the second half of the month of June 1936, following Dr. Aston's visit to Tokyo, we had three more lectures by Professor Kimura: on Wednesday June 17, on Wednesday June 24 and on Friday June 26, before the summer recess started at the beginning of the month of July. The two-hour lecture on June 17 covered the principles of x-ray diffraction, Bragg's experiment, followed by Debye and Scherrer's experiment. The two-hour lectures given on June 24 and 26 were detailed explanations of the properties of X-rays, designs of x-ray tubes and the Siegbahn-type x-ray spectrograph, which was placed in a room across from Professor Kimura's office. The final lectures given on June 24 and 26 were two of the most memorable lectures I have ever attended in my life (See Fig.3 and Fig.4). They were highlighted by an extremely detailed explanation of Niels Bohr's theory on the structure of the atom. I was particularly intrigued by a remark made by Professor Kimura that he and Dr. Yoshio Nishina attempted to discover element 93 by the use of the Siegbahn-type x-ray spectrograph at the Niels Bohr Institute in Copenhagen, Denmark as early as in 1925. It seemed to me

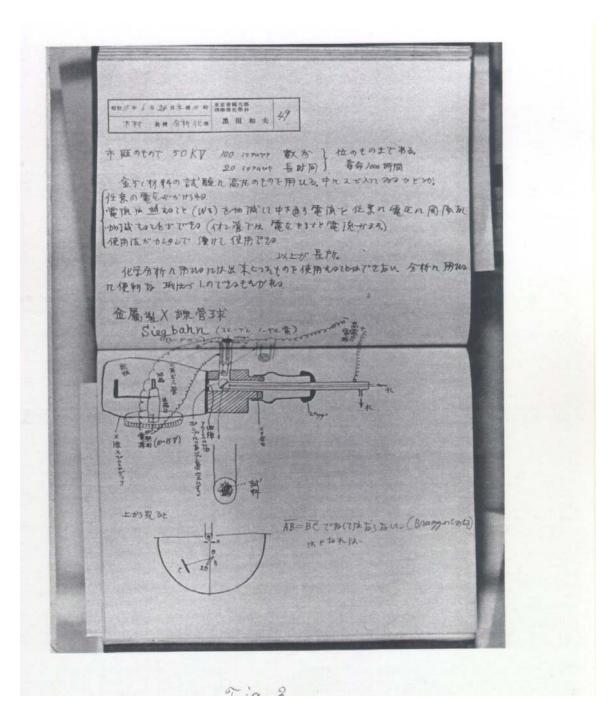


Figure 3. This is a copy of Kuroda's notes from Professor Kimura's introductory lecture on Niels Bohr's theory on the structure of the atom on June 24, 1936.

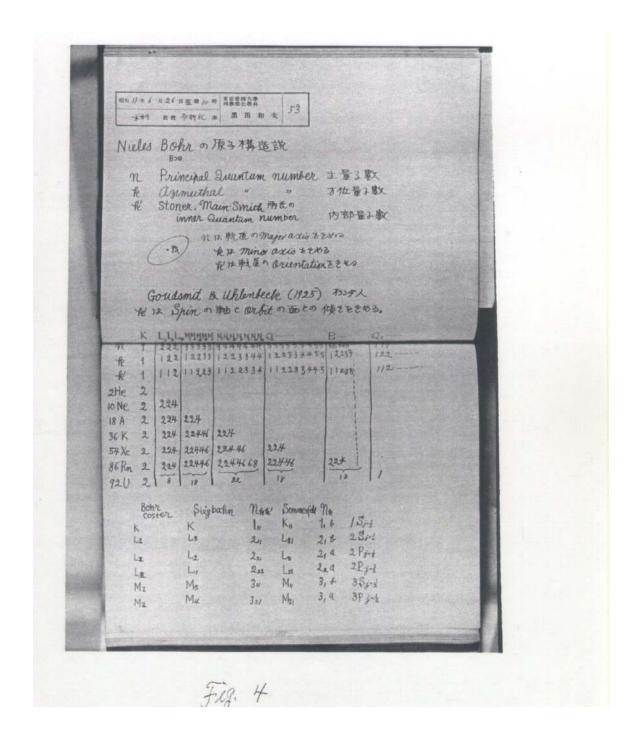


Figure 4. This is a copy of Kuroda's notes from Professor Kimura's second lecture on Niels Bohr's theory on the structure of the atom on June 26, 1936.

that the x-ray spectrograph was the most powerful tool to be used for finding a new element, and the mass-spectrograph invented by Dr. Aston must be the most important tool in chemistry perhaps in some distant future.

By the time the final lecture of the Spring Semester was given on Friday June 26, 1936, I was still reading Dr. Aston's book in the evenings at home. The latter part of the book became increasingly difficult and there were many English words which I did not know. Chinese characters and pencil marks I left here and there 55 years ago indicate that I was finally able to reach the final chapter of the book by the end of July 1936.

At about the time Dr. Aston was visiting Tokyo in June 1936, a 31-year-old physicist from Italy was spending the summer touring the United States. His name was Emilio Segrè and he was the first PhD student of Professor Enrico Fermi at the University of Rome. While touring the United States, he visited the University of California at Berkeley, where the 37-inch cyclotron built by Professor Lawrence was in full operation. Both Fermi and Segrè had been keenly interested in the cyclotron, but it appeared to them that to acquire this machine was beyond their technical and financial capabilities. Italy, in the 1930's, was considered as one of the five most powerful nations in the world, but not one of the richest.

When Segrè visited Berkeley in the summer of 1936, he obtained from Lawrence several scraps of molybdenum deflectors that were quite radioactive. He planned to study them on his return to Palermo, treating them as though they were an ore. The return journey from California to the Island of Sicily by train and ship in those days must have taken at least a month, so that any short-lived isotopes with half-lives less than several days must have completely decayed away during the journey. Since Segrè was a nuclear physicist and not a radio-analytical chemist, he asked the help of a professor of Mineralogy, named Carlo Perrier, to improve his chemical prowess. Segrè and Perrier

had become close friends when the former arrived at Palermo from Rome one year earlier in 1935.

While Segrè was beginning his qualitative analyses of the samples of the "ore" which he had brought back from California, at the University of Palermo in the fall of 1936, the first of Professor Kimura's lectures in analytical chemistry at the Imperial University of Tokyo began at the regularly scheduled time at 10 a.m. on Wednesday September 16, 1936. On that day, he began his lecture series in quantitative analysis, which was to last from September 16, 1936, through February 10, 1937, with the subject of volumetric analysis. He wrote on the blackboard the titles of three important reference They were Classen's "Theorie und Praxis der Massanalyse", Sutton's "A Systematic Handbook of Volumetric Analysis", and I. M. Kolthoff's "Die Massanalyse". The series of lectures on the subject of volumetric analysis lasted until November 25, 1936, and then Professor Kimura began to cover the subjects of gravimetric analysis and gas analysis, which ended on December 2, 1936. On Wednesday January 27 he began his lectures on colorimetric analysis and continued on to discuss nephelometric analysis. On Wednesday February 3rd he lectured on quantitative spectroscopic analysis and quantitative analysis by means of the x-ray spectrographic method. He taught us in detail how to determine accurately the ratios of hafnium to zirconium in minerals by adding known amounts of lutetium oxide (Lu_2O_3) and comparing the intensities of the L β_2 x-ray line (1367 x-ray unit) and L β_1 x-ray line (1371 x-ray unit). Professor Kimura was an extremely polite and modest gentleman, but he casually remarked on this day that this procedure was what Professor Hevesy used to refer to as the "Kimura's method" of quantitative x-ray spectrographic analysis.

There were one or two more lectures delivered by him during the month of February 1937, but unfortunately I failed to write down the dates in my notebook. The last topic covered was an outline of geochemistry and each student received a total of no less than eighteen mimeographed tables of abundances of elements in the earth's crust, average compositions of the atmosphere, igneous rocks, volcanic rocks, volcanic gases, hot springs, and so on. It would have been an enormous task to memorize everything, which was taught in this course, which began in April 1936 and lasted until February

1937, in preparation for the final written examination scheduled during the month of March 1937. We were greatly relieved however, when Professor Kimura announced that the final examination would be substituted by the presentation of a written essay on the analytical chemistry of an element to be submitted by him. The element to which I was assigned was titanium. Thanks to the extreme generosity of Professor Kimura, all of us who attended his lectures thus received the grade of "excellent" in analytical chemistry at the end of March 1937.

2. The Twelfth Year of Showa: 1937

The new school year started on April 1, 1937, and it was cherry blossom time again in Tokyo. Then, one day, a notice was placed on the bulletin board at the main entrance of the Chemistry Building of the Tokyo Imperial University. It was the announcement that Professor Niels Bohr from Copenhagen, Denmark, will be visiting the university and giving a lecture series on the structure of the atom, beginning on April 23, 1937. I knew that Professor Bohr was the recipient of the 1922 Nobel Prize in Physics "for his services in the investigation on the structure of atoms, and of the radiation emanating from them". Besides, a decade earlier Professor Kimura had spent two years in Copenhagen working under Niels Bohr and George de Hevesy. As I mentioned earlier, Professor Kimura lectured on Bohr's theory on the structure of the atom on June 26, 1936, but now I was to have the great fortune of attending a series of lectures on the subject to be given by Professor Bohr himself. I therefore went to the Maruzen bookstore and purchased the second edition of "The Theory of Spectra and Atomic- Constitution" by Niels Bohr, published by the Cambridge University Press in 1924. The price of the book was 6 Yen and 30 Sen, which was exactly one-half of 12 Yen and 60 Sen which I had paid for "Mass-Spectra and Isotopes" by F. W. Aston ten months earlier.

Niels Bohr's lecture was much more difficult for me to understand than the lecture given by Francis Aston ten months earlier. After all, I was a 20-year-old chemistry student in 1937 and I felt that whatever Bohr was telling us on that day must be absolutely correct. I was particularly interested in the new periodic table which appeared in the 1924 edition of his book. It had five vacancies between element 1, hydrogen, and element 92, uranium. They were elements 43, 72, 75, 85 and 87. In a table which appears in the Appendix section of this book, however, element 72 is listed as Hf. The reason for this is that the first edition of Bohr's book was published in 1922 and the book I purchased in 1937 was the second edition published in 1924, the main part of which

was the same as the 1922 edition but the APPENDIX section was added to update the 1922 edition.

Meanwhile, element 72 was discovered in January 1923 by Coster and Hevesy who were both working in Bohr's laboratories in Copenhagen. Niels Bohr's theory predicted that the missing element 72 should have chemical properties similar to those of zirconium; Coster and Hevesy found this element in zirconium minerals, while other scientists were looking for it in vain, assuming that it belonged to the rare earths. The discovery of hafnium by Coster and Hevesy in January 1923, which was announced one month after Bohr received the 1922 Nobel Prize in Physics and delivered his Nobel Lecture entitled "The structure of the atom" on December 11, 1922, was a dramatic confirmation of the correctness of Bohr's theory concerning the structure of the atom. The discovery of hafnium also demonstrated the extreme usefulness of the x-ray spectrographic method of analysis in finding "missing" elements. In fact, two years later in May 1925, Walter Noddack and his assistants Ida Tacke and Otto Berg at the University of Berlin announced the discoveries of two of the remaining missing elements: element 75 rhenium and element 43 masurium. They also used the x-ray spectrographic method to detect the presence of these elements. It so happened however, that while Noddack and co-workers were soon able to enrich and isolate element 75 rhenium from various minerals and ores, they failed to enrich element 43 masurium, and the new element masurium which they reported to have discovered in 1925 was generally regarded as questionable twelve years later, in 1937.

Meanwhile, at the University of Palermo on the island of Sicily, Italy, Perrier and Segrè published three papers in the *Journal of Physical Chemistry* and in *Nature*, in which they reported that the molybdenum scraps which Segrè brought back to Italy from Berkeley in 1936 exhibited a strong unknown radioactivity. A careful study of this new radioactivity led them to the conclusion that element 43 is formed by the nuclear reactions

94
Mo(d,n) 95 ₄₃ (60-day half-life)

and

With unweighable amounts, observations were made on the chemical behavior of the new element. What they did was essentially qualitative analysis based on the concept of radioactive indicator, which had been invented by my teacher's teacher, George de Hevesy and his co-worker F. A. Paneth as early as in 1913.

Element 43 was the first new element to be discovered by means of artificial synthesis, and this discovery marked the beginning of a new era in the history of chemistry. This important discovery was not immediately accepted by all the chemists of the 1930's. For example, in the 1941 edition of the Periodic Table in "General Chemistry for Colleges" by B. Smith Hopkins, published in the United States in 1942, element 43 is shown by the symbol "Ma", which was the name "masurium" proposed by Noddack and co-workers in 1925.

It was ten years later in 1947, that the discovery by Perrier and Segrê became universally accepted by the scientific community, and the name "technetium" with the symbol Tc which they proposed for element 43 became official.

The construction of the 26-inch Tokyo cyclotron was nearing completion and undergoing tests in 1936. I had a number of occasions to visit the cyclotron laboratory located at the Institute of Physico-Chemical Research with some of my classmates and marvel at this magnificent machine. The cyclotron, which undoubtedly cost a large sum of money, and which did not exist in Italy in 1936, was undergoing tests. While I was attending Professor Kimura's lectures in analytical chemistry during the period between April 1936 and February 1937, all of the twenty-four first-year chemistry-major students each had a laboratory desk space assigned and had access to all the glassware and reagents needed for qualitative and quantitative analyses including such expensive items as platinum crucibles. I could have easily asked Dr. Nishina if he had a few scraps of molybdenum deflectors. I could have brought back the irradiated molybdenum samples to the Chemistry Building and I could have carefully examined the chemical properties of the radioactive isotopes in them, just as the young Italian physicist Emilio Segrè was

doing in his laboratories at the University of Palermo in Italy. The 26-inch Tokyo cyclotron was smaller than the 37-inch Berkeley cyclotron and hence the beam intensity of the Tokyo cyclotron may not have been as high as that from the Berkeley cyclotron. But, I had an advantage over Segrè, because it took him at least a month to bring back his samples from Berkeley, California, to his laboratories in Italy by train and ship, whereas the distance between the Institute of Physico-Chemical Research to the Chemistry Building of the Tokyo Imperial University was only a mile or two and could be reached in about ten to twenty minutes.

3. The Thirteenth Year of Showa: 1938

In March 1938, the students who had entered the University in April 1936 had completed all the required courses in the Chemistry Departments. The required coursework consisted of one year of analytical chemistry taught by Professor Kenjiro Kimura, two years of physical chemistry taught one year each by Professor Jitsusaburo Sameshima and Professor Masao Katayama, one year of electrochemistry taught by Professor Yohei Yamaguchi, two years of inorganic chemistry taught by Professor Yuji Shibata, and two years of organic chemistry taught by Professor Bennosuke Kubota. Each student was to choose his major professor under whom he was to carry out research work. I wanted to make sure to preserve the lecture notes of all these professors for the future. So I went to a book-binder's shop and asked them to bind them each in the form of a German textbook. All of them were lost during and after World War II, except for the notes of Professor Kimura's lectures (See Fig.5).

Several days before the new school year was to begin on April 1, 1938, I went to Professor Kimura's office and asked him to be my major professor. The thesis problem which he gave me on that day was to study the distribution of ²²⁶Ra among associated minerals occurring in the pegmatite regions at Yamano-o, Ibaragi Prefecture, and at Teshirogi, Fukushima Prefecture. I was somewhat surprised at his choice of thesis problem, because the 26-inch Tokyo cyclotron had become operational in 1937 and a joint project by the groups of Dr. Nishina and Professor Kimura on the irradiation of uranium and thorium with fast neutrons was in progress by the spring of 1938. Although I tried not to show any sign of disappointment with the research problem given to me by Professor Kimura, he may have sensed what I really had in my mind, as he went on to tell me that one of the most important requirements for a doctor's dissertation was that it had to be based on the candidate's own research, and joint papers published with physicists were not acceptable as the main part of one's doctoral dissertation. Another important requirement was that a doctor's dissertation had to be written not in Japanese, but in one of the following three foreign languages: English, German or French.

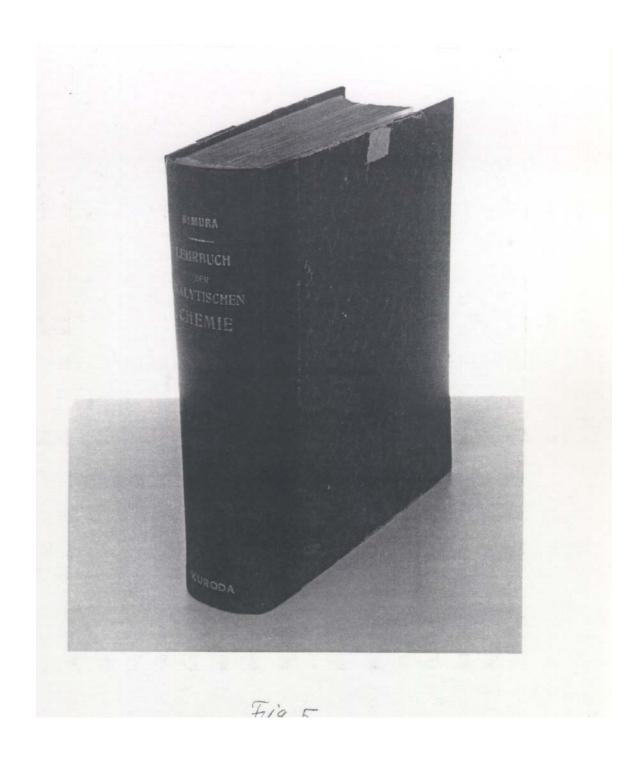


Figure 5. This is Professor Kimura's lecture notes, *Lehrbuch der Analytischen Chemie*, that Kuroda had bound in 1938. Pages from this were reproduced as Figs. 1-4 and 8.

I did not understand why Professor Kimura seemed to be so much concerned about the matter of joint publications with physicists at that time; however, it soon became clear to me when the manuscript submitted by the Nishina-Kimura group began to appear in the British journal *Nature* and in the American journal *Physical Review*. The first paper published in *Nature* in 1938 entitled "Artificial production of uranium Y from thorium" was co-authored by Nishina, Yasaki, Ezoe and Kimura. The next four papers which appeared in 1939 and 1940 were co-authored by Nishina, Yasaki, Ezoe, Kimura and Ikawa. The first three authors were physicists and the last two were chemists. Professor Kimura had hired a retired analytical chemist named Masao Ikawa in 1936 as his technical assistant in anticipation of the forthcoming joined research on uranium and thorium. Since the 26-inch Tokyo cyclotron became operational in 1937, Mr. Ikawa became in charge of purification and preparation of all the uranium and thorium samples to be irradiated with fast neutrons as well as of the isolation and final purification of all the carriers added to uranium and thorium. The purified samples were to be returned to the physicists who did all the radioactivity measurements. In short, chemists played a supporting role. They took orders from physicists and the relationship between them was that of a master and a servant.

It is worthy of note here, however, that in Europe and the U.S. during the period from 1938 to 1940, chemists rather than physicists played a key role in making most of the important scientific discoveries. It was two radiochemists in Germany, named Otto Hahn and F. Strassmann, who discovered fission. The transuranium elements neptunium and plutonium were discovered in 1940, by McMillan and Abelson (element 93) and Seaborg, McMillan, Kennedy and Wahl (element 94). McMillan was the only physicist among those mentioned above. After the Japanese attack on Pearl Harbor on December 7, 1941, however, physicists regained the leadership. The first nuclear reactor was built at the University of Chicago by Enrico Fermi, who was a physicist, and his assistants. The atom bombs were built during a period of eighteen months by a large number of scientists working at Los Alamos, New Mexico, during the period from 1943 to 1945, under the leadership of Robert Oppenheimer, who was a young physicist from the University of California at Berkeley.

It seems to me that there were a number of reasons why Professor Kimura gave me the thesis problem to measure the radium contents of minerals, instead of assigning me to work on the cyclotron project. First of all, being a Christian, he was not at all interested in such ideas as building atom bombs and dropping them over the cities of enemy countries in order to kill a large number of innocent people. Secondly, after spending two years in Copenhagen and studying under the guidance of such eminent scientists as Niels Bohr and George de Hevesy, he knew how far Japan was behind Europe and the United States in science. There was no chance of winning if Japan had challenged these highly advanced countries. The scientists in Japan at that time should be making a long-range plan for the future, regardless of the outcome of a possible war against the United States and the rest of the free countries.

The third reason I could think of, and this may be the most important of all, was that Professor Kimura may have thought that if this new student named Kuroda was a clever student and truly dedicated to science, and if he and Mr. Ikawa worked together in a new laboratory space which had just been completed at that time between the old and the new Chemistry Buildings, next to the brand-new office of Professor Shibata, they, Mr. Ikawa and Kuroda, the oldest and the youngest analytical chemists, just might accidentally come up with a big surprise, which might turn out to be an extremely important scientific discovery.

So, Mr. Ikawa and I began working in the brand-new laboratory every day. He was working on the purification of uranium and thorium samples to be irradiated at Dr. Nishina's cyclotron laboratories, which then came back to him for the chemical separations and purifications of the various fractions. These were then sent to the Physics Building for the measurements of the radioactivities. Mr. Ikawa was a very hard-working elderly gentleman. He seldom spoke to me and I decided not to disturb him by asking stupid questions while he was doing his experiments. I could tell whether his experiment was going well or not by a loud sound which came out of his mouth now and then. His samples of irradiated uranium and thorium must have been loaded with fission products

and highly radioactive. It never occurred to me however, to ask him how radioactive those irradiated uranium and thorium samples were.

Mr. Toshio Nakai and Mr. Hiroshi Hamaguchi, who graduated from the University a few years earlier than I, were kind enough to teach me how to carry out the analyses of radium in detail. My job was to first grind up twenty grams of minerals to a fine powder in an agate mortar and then carry out the sodium carbonate fusion in a platinum crucible. The melt was dissolved in nitric acid, and insoluble silica was removed by treating with sulfuric acid and hydrochloric acid. When all the insoluble residues were brought into solution, it was sealed in a "Curie bottle" and stored for one month until the radioactive equilibrium was established between the parent ²²⁶Ra and the daughter ²²²Rn.

The sample preparation, especially grinding up twenty grams of large crystals of mica to a fine powder, was an extremely tedious operation and required a few days of hard labor. Converting all of the powdered sample into a clear solution to be stored in a "Curie bottle" could be done in one day, but occasionally the solution became cloudy and insoluble residues formed, in which case the carbonate fusion process had to be repeated. The measurement of radioactivity was also time-consuming. After setting aside the "Curie bottle" for 30 days, it had to be connected to a vacuum system and ²²²Rn had to be quantitatively transferred into an ionization chamber. The sample solution in the "Curie bottle" was placed over a water bath and the solution was gently boiled in vacuum. After all the radon gas was transferred into the ionization chamber, one had to wait for several hours until the transient radioactive equilibrium between ²²²Rn and its short-lived decay products RaA, RaB, RaC etc. was established. The velocity of the movement of a gold leaf inside the ionization chamber had to be measured through a microscope. The radium content of a mineral as low as 0.02 pCi/g or 2xl0⁻¹⁴ grams per gram could be determined by taking a 20-gram sample. By working hard six days a week, I was able to determine the radium contents of a total of 15 samples of pegmatite minerals in two months. At this rate, I could have analyzed ninety samples in a year.

While I was doing the radium analyses during the months of April and May of 1938 however, a thought occurred to me that what I had been doing was to determine the uranium contents of the minerals, assuming that ²³⁸U and ²²⁶Ra in these minerals were in a secular equilibrium. In order to do this, I was first separating ²²⁶Ra from the mineral and waiting 30 days until a secular equilibrium between the parent ²²⁶Ra and the daughter was established. Even when I determined the amount of which was introduced into the ionization chamber, I had to wait several hours until a transient equilibrium between ²²²Rn and its daughters was established. On top of all this waiting, I had to observe the exact position of the gold-leaf through a microscope once every hour for several hours. A great deal of waiting was involved to determine the content of ²²⁶Ra in a sample of mineral according to this old-fashioned method, even though I could have been doing other things during these waiting periods. Moreover, I was not absolutely sure that the initial separation of ²²⁶Ra from the mineral sample was quantitative, and the recovery of ²²²Rn from the ²²⁶Ra solution may not have been 100 percent, even though I was extremely careful in performing each step of the procedures.

I felt that it would be very interesting to determine the amounts of other isotopes of radium in these minerals. In 1938, it was known that there were four isotopes of radium, namely ²²³Ra(AcX), ²²⁴Ra(ThX), ²²⁶Ra and ²²⁸Ra(MsThI). Two of these isotopes, ²²⁴Ra and ²²⁸Ra belonged to the ²³²Th decay chain, while ²²³Ra belonged to the ²³⁵U decay chain, although this had not been discovered until a young physicist named Alfred Nier at the University of Minnesota succeeded in identifying this important isotope of uranium in 1939. One of the two isotopes of radium, ²²⁴Ra and ²²⁸Ra could be used as a tracer or a "radioactive indicator" to calculate the chemical yields of other radium isotopes, taking advantage of the concept of radioactive tracer published by George de Hevesy and Fritz Paneth in a paper entitled "RaD als 'Indikator' des Bleis" (RaD as indicator of lead) in *Zeitschrift der anorganischen Chemie* **82** (1913) 322. If the contents of ²²⁶Ra and ²²⁴Ra or ²²⁸Ra in these minerals could be accurately determined, the contents of not only ²³⁸U but also ²³²Th could be calculated. It would be very interesting to find out the uranium to thorium ratios of these minerals by developing a method based on the determination of the contents of radium isotopes. While I was thinking about

experimenting with ²²⁴Ra and ²²⁸Ra to test these ideas, I received a new assignment from Professor Kimura, which caused me to postpone the performance of these experiments temporarily.

At about this time in Europe, two groups of scientists were trying to identify the radioactive nuclides produced when uranium was bombarded with slow neutrons from radon-beryllium sources, which were similar to the source being used by Fermi and coworkers since 1934. These sources emitted about $\approx 10^7$ neutrons per second. In the summer of 1938, there were only two cyclotrons in operation in the world: one was the 37-inch machine at Berkeley in California and the other was the 26-inch machine in Tokyo, Japan.

In Paris, on July 12, 1938, Irene Curie and Pavel Savitch reported to have discovered a 3.5-hour radioactive component, which chemically resembled lanthanum, in neutron-irradiated uranium. They did not realize at that time that it was indeed ¹⁴¹La, which is a fission product. In Berlin, meanwhile, Hahn, Meitner and Strassmann were also attempting to identify the radioactive isotopes found in irradiated uranium samples, but they thought that these isotopes were trans-uranium elements, Eka Re, Eka Os, Eka Pt, etc. and they were trying to separate them from each other by adding the carriers Re, Os, Pt, etc. to the irradiated sample, just like Mr. Ikawa was doing at the University of Tokyo. By November 8, 1938, Hahn and co-workers had changed their mind after reading the paper by Curie and Savitch, and they reported that most of the activities were due to the isotopes of radium and its decay products, actinium and thorium, as shown below:

"Ra I"?
$$-\beta$$
-> Ac I $-\beta$ -> Th? <1Min <30 Min
"Ra II"? $-\beta$ -> Ac II $-\beta$ -> Th?
$$14 \pm 2 \text{ Min.} \approx 2.5 \text{ Hours}$$

"Ra III"?
$$-\beta$$
-> Ac III $-\beta$ -> Th? 86 ± 6 Min many days "Ra IV"? $-\beta$ -> Ac IV $-\beta$ -> Th? $250-300$ Hr 40 Hours

(Dr. Kuroda's original drawing of these decay equations is reproduced in the Appendix.)

Early in December 1938, they thought that they had established the above decay chains in which the genetic relations appeared to be solidly known. Supposedly four isotopes of radium were decaying to Ac and then to Th. To make doubly sure, however, Hahn and Strassmann decided to identify the radium isotopes beyond any doubt. They submitted them to several tests using barium as a carrier and ²²⁸Ra(MsTh1) as a radioactive indicator. These experiments which were carried out by Hahn and Meitner in December 1938 forced them to conclude that the hypothetical radium isotopes were in fact barium! This was the way the important discovery of nuclear fission was made by two German radiochemists, Professors Otto Hahn and Fritz Strassmann. They did not have access to expensive machines such as the cyclotron. The equipment which they used was a source of neutrons made of radon and beryllium, and a primitive counter.

As I mentioned earlier, in the spring of 1938, I was thinking about using ²²⁸Ra(MsTh1) as a radioactive indicator. In the small laboratory which I shared with Mr. Ikawa, samples of neutron-irradiated uranium and thorium were arriving all through the year 1938. In those days I did not know how radioactive these samples were, but judging from the huge amounts of uranium and thorium compounds which Mr. Ikawa was purifying almost daily, and because the neutron flux from the 26-inch cyclotron must have been far greater than that from the radon-beryllium source used by Hahn and Strassmann in Germany, the irradiated samples which were delivered to Mr. Ikawa must have been highly radioactive and loaded with Ra I, II, III, and IV. I could have easily asked Mr. Ikawa to spare a small fraction of these samples to test my idea. I could have

performed these experiments six months earlier than Hahn and Strassmann and tell Professor Kimura that these samples contained radioactive barium isotopes. The research problem which I was given by Professor Kimura in March 1938 could have led to an extremely important scientific discovery. Unfortunately, before I learned about all these exciting discoveries being made in Paris and Berlin, I was assigned to a new project which was being initiated by Professor Kimura.

One day in late spring of 1938, Professor Kimura told me: "Mr. Kuroda, how would you like to spend the summer in Hakone Hot Springs"? Apparently, it was an extension of the project initiated in the early 1930's by Professor Yuji Shibata entitled "Geochemical Investigation of Volcanoes in Japan". Hakone was one of the most famous spas in Japan, where many of the wealthy families, including Emperor Hirohito of Japan, used to spend the summer in their villas. There was no way I could have declined the offer to spend a summer in Hakone, all expenses paid by the University. So, I told my professor that of course I would be delighted to do so. It turned out that my new assignment was to read a paper which had been recently published by Madame Ida Noddack, entitled "Uber die Allgegenwart der chemischen Elemente", which appeared in the German journal Angewandte Chemie 47, page 835 (1936). In this paper, she argued that with sufficient labor and time, one should be able to find all the chemical elements in a sample of minerals. For example, she found in a sample of zincblende from Mansfeld all except 16 elements out of 89 known elements. The analyses of not only the natural minerals, but also of organic substances and artificial products revealed the same trend: the greater the detection sensitivity, the greater the number of chemical elements detected in the sample. What Professor Kimura wanted me to do was to test the hypothesis of Ida Noddack by analyzing the water of hot springs, because she did not mention if the same rule applied to natural waters. My first reaction to this assignment was that it would take at least six months, but Professor Kimura also told me that I should visit the hot springs perhaps once a month for a period of at least one year and observe the seasonal variations of temperature, pH, amount of flow and chemical compositions. This meant that I would be tied up with the hot springs project most likely for one year or even much longer.

I did not know it until many years later, but while I was planning for the forthcoming expedition to Hakone Hot Springs, Emilio Segrè at the University of Palermo in Italy was planning to visit Berkeley to search for the short-lived isotopes of element 43, which could not stand the duration of a sea trip from California to Italy. In the early summer of 1938, he purchased a roundtrip ticket from Palermo to Berkeley and left Italy for the United States. By the time he reached Berkeley however, he learned that he lost his professorship at the University of Palermo, because Mussolini had embraced anti-Semitism and all Jewish state employees were being fired. So he had to stay in America, being separated from his family in Italy. Fortunately, however, he received a temporary research position at Berkeley and soon became acquainted with a young American chemist named Glenn T. Seaborg, who had just received his PhD degree from the University of California. Seaborg and Segrè worked together on short-lived isotopes of element 43 and they soon discovered a new isotope, which is now known as ^{99m}Tc. They did not anticipate it at that time, but this isotope was to become one of the mainstays of nuclear medicine. Seaborg was five years older than I and he was 26 years old then. If I had been a chemist as bright as Seaborg in 1938, I should have suggested to Professor Kimura in the early spring of 1938 that we should start looking for short-lived isotopes of element 43, using the 26-inch Tokyo cyclotron, but of course, I did not do so.

I first visited Hakone on July 20, 1938, to survey the area where a group of hot springs is located. They were all acid springs with pH about 2 and rich in hydrogen sulfide. They were located near the highest peaks of Hakone volcano. It was a beautiful place to spend a summer, but I had to carry a heavy load of instruments and chemicals on my back and walk a mile or two from the nearest bus-stop. I stayed at Yunohanazawa from August 10 till September 1, 1938, doing daily field measurements. Then I continued going there once every month until the summer of 1939, when I stayed at Yunohanazawa from July 31 to September 9, 1939.

Optical spectrographic analyses of the water of Hot Springs revealed the presence of the following trace metals: Ti, V, Mn, Zn, Ge, Sb, Bi, Cr, Ni, and Co. According to the gravimetric method, I carried out the analyses of the major constituents of three

samples of hot springs: Na, Ca, Mg, Fe, Mn, Al, Cl, HPO₄, SO₄, HSO₄, HBO₂, H₂SiO₃, H₂S, and CO₂. Then I began the analyses of trace metals by the polarographic method and by the colorimetric method by the use of dithizone. The analyses of strontium and barium were carried out gravimetrically strictly according to the method described on page 750 of the 1929 edition of Hillebrand and Lundell's "Applied Inorganic Analysis". Mixed nitrates of strontium and calcium were treated with a mixture of equal volumes of alcohol and ether. I found that a sample of water contained 10 micrograms of barium and 32 micrograms of strontium per liter.

In the late fall of 1938, I was beginning to feel that all these analyses of water may be important, but I was just following the procedures which were recommended by other analytical chemists and I would never make an exciting discovery from this kind of research. Then in the late fall of 1938, I learned that Enrico Fermi and Richard Kuhn were to be awarded the 1938 Nobel Prizes in Physics and Chemistry, respectively. At the beginning of 1939, a notice appeared on the bulletin board of the Chemistry Building announcing the availability of an Italy-Japan exchange fellowship for interested students. The instant I saw this notice, I decided to take the examination. I intended to go to the University of Rome and continue my research in the laboratories of Professor Enrico Fermi, where I could possibly work under Emilio Segrè, whom I thought was a radiochemist. I had enough money in my bank account to purchase a one-way ticket from Tokyo to Rome, which was 90 Yen or about 45 U.S. dollars. I immediately went to a bookstore in front of the main gate of the University and purchased a book entitled "Italian in Three Weeks" and began studying the Italian language. Italian was not taught in most schools in Japan and I knew that none of the professors and students in the Faculty of Science of Tokyo Imperial University had a good knowledge of the Italian language, but I was hoping I could pass the examination by studying the language intensively in the short time I had. I failed to pass the examination, however, because it was given in French instead of Italian! Although I did not know it at that time, both Fermi and Segrè were in the United States, because of the political situations which prevailed in Europe in the late 1930's.

There used to be an old saying in Japan that "Man lives only fifty years". I never met Professor Fermi in my lifetime, because he lived for only 53 years. In 1954 he became seriously ill in Chicago, while I was working hard on the theory of the Pre-Fermi natural reactor (P. K. Kuroda, *J. Phys. Chem.* 25, 781 (1956) at Fayetteville, Arkansas. I met Professor Segrè only once in my life. It was on October 12, 1988, exactly 50 years after 1938, when I was planning to work under him in Italy. He visited me in Las Vegas on that day, which happened to be the very day Professor Kimura passed away at his home in Yokohama.

4. Prelude to War: 1939—1940

Soon after the news of the discovery of fission reached Japan in early 1939, Mr. Ikawa became overwhelmed by his work of separating the fission products from uranium irradiated with fast neutrons. Two new students who joined our group in April 1939 were therefore assigned by Professor Kimura to work together with Mr. Ikawa. Their names were Mr. Nobufusa Saito and Mr. Jiro Matsuura.

Three papers co-authored by Nishina, Yasaki, Ezoe, Kimura and Ikawa were published in 1939 and 1940 in the British journal *Nature* and the American journal *Physical Review*, announcing the important discovery of symmetric fission of uranium and thorium. Another important paper by Nishina, Yasaki, Ezoe, Kimura and Ikawa appeared in the American journal *Physical Review* in 1940, announcing the discovery of a new isotope of uranium with a half- life of 6.5 days. They stated that this must be ²³⁷U produced from ²³⁸U through the loss of a neutron, as in the case of the production of UY (²³¹U) from thorium (²³²Th), which was reported by Nishina, Yasaki, Ezoe and Kimura in 1938 in *Physical Review*. The discovery of ²³⁷U which beta-decayed with a half-life of 6.5 days was extremely important, because the daughter had to be the then-unknown transuranium element with the atomic number 93.

To search for element 93, the irradiated uranium oxide was left for about 7 days, and was then dissolved in nitric acid. The solution was then treated with ammonium sulfide after the addition of perrhenic acid. The precipitated rhenium sulfide was examined for beta- and alpha-activities. Neither of them could be found and it was concluded that it decayed into a very long-lived element 93.

The discovery of symmetric fission by the Tokyo cyclotron group was immediately confirmed by Segrè and Seaborg in a paper submitted to the *Physical Review* on December 12, 1940, and it appeared in the 15 January 1941 issue. This was

the last report from the investigators in the United States to reach Japan and it created an impression among the Japanese investigators that the Tokyo cyclotron group was ahead of the Berkeley cyclotron group, because the American scientists were repeating the discovery made by the Japanese scientists.

At this time at Berkeley, however, the powerful 60-inch cyclotron was in operation, while the Japanese scientists were still depending on the old 26-inch cyclotron. It was obvious that Japan needed to speed up the construction of a 60-inch cyclotron to keep up with the competition of the Berkeley group. Dr. Ryokichi Sagane, who was the son of the President of the Japanese Imperial Academy of Science, Professor Hantaro Nagaoka, was visiting the University of California in March 1939. Sagane asked Professor Lawrence for the blueprint of the 60-inch cyclotron, but his request was denied. So the Tokyo group had to build the 60-inch cyclotron on its own without having access to the blueprint of the Berkeley cyclotron.

In the paper which appeared in the 15 January 1941 issue of the *Physical Review*, Segrè and Seaborg described that they irradiated uranium and thorium with 17-MeV neutrons (16-MeV deuterons on beryllium) from the 60-inch Berkeley cyclotron. In early 1941, Japanese investigators had no way of knowing that the Berkeley group was bombarding uranium with deuterons and discovering the important isotopes of element 94: ²³⁸Pu and ²³⁹Pu. It was highly significant that a manuscript entitled "Radioactive element 94 from deuterons on uranium" by Seaborg and co-workers, which was submitted on January 28, 1941, did not appear until five years later in the April 1, 1946 issue of *Physical Review*. The investigators in Tokyo were not aware of the fact that they had been soundly beaten by American scientists in the competition for discovering new elements heavier than uranium, almost a year before Pearl Harbor.

Meanwhile, I graduated from the University in March 1939 and received the degree of "Rigaku-shi", which was roughly equivalent to the Master of Science degree in the United States. I was told by Professor Kimura to continue my work on trace elements in the water of Hakone Hot Springs. In the early summer of 1939 however, he gave me a

new project, which was to begin a study of the famous radioactive springs at Masutomi, Yamanashi Prefecture, located about 60 miles west of Tokyo near Mount Fuji. In July 1939, Professor Kimura, Mr. Shinya Oana and I made a short visit to Masutomi. Mr. Oana had graduated from the University one year earlier; he was an assistant of Professor Yuji Shibata and he was to measure the "heavy water" contents of Masutomi Springs, while I did the measurements of radium and radon contents. I felt that the study of Masutomi was far more exciting than that of Hakone. The radon concentrations of some of the springs were on the order of a few tenths of microcurie (10⁻⁶ Curie) per liter and were known to be the highest among all the hot springs and mineral springs in Japan.

Soon after the discovery of fission, scientists in Europe were beginning to create a uranium chain reaction using heavy water as a moderator.

Masutomi was a small village deep in the mountains, opposite from Mount Fuji to the south, along the gorges of the Hontani River, totally isolated from the outside world. There was no transportation beyond another village five miles away, where the bus stop was located. We had to walk the distance from the bus stop to the best hotel in Masutomi, owned by Mr. Tanehito Tsugane. Professor Kimura was treated there as if he was a lord visiting a small villa with two of his favorite retainers.

Mr. Tsugane's mother once showed me a very old impressive-looking document, which described the family tree which went back to the 16th century. Their ancestor was one of the retainers of Lord Takeda-Shingen, who was at that time generally regarded as one of the most likely candidates to become a shogun, which was a military governor of Japan.

The episode of the untimely death of Lord Takeda was introduced to the western world by Akira Kurosawa's popular movie "Kagemusha" in the 1970's. Recently, Mr. Kurosawa received a special Academy Award in Hollywood, California, and his latest film "The Dreams of Akira Kurosawa" was showing in a movie theater in Las Vegas, Nevada. In one of the episodes of this movie, there is a scene of Mount Fuji collapsing

after being engulfed in a nuclear fire, which was triggered by an uncontrollable nuclear chain reaction. This reminded me of the old days when Mr. Oana and I were measuring the heavy water contents and the radioactivities of the water of Masutomi Springs a half-century ago.

I did not understand why Professor Kimura did not order me to join the group of Mr. Ikawa and assist him in chemical separations and identifications of fission products found in irradiated uranium, but it seems to me that he already knew that the joint project with Dr. Nishina's cyclotron group was nearing the end, because the construction of the 60-inch cyclotron was progressing at an unusually slow rate and without it, Japanese investigators would never be able to compete successfully with American scientists. Come to think of it now, it is quite possible that he was hoping that Mr. Oana and I might find some radioactive fission products in the Masutomi Radium Springs produced by "natural" fission of uranium atoms in a uranium ore deposit, which might exist deep in the interior of the earth in the vicinity of Masutomi, since this region was known for the existence of gold mines from the days of Lord Takeda.

It is significant that the Tokyo cyclotron group published three important papers in the British journal *Nature* and the American journal *Physical Review* in 1939 and 1940, whereas no paper was published during the year 1941.

On January 9, 1942, Nishina, Kimura, Yasaki and Ikawa submitted a manuscript entitled "Einige Spaltprodukte aus der Bestrahlung des Urans mit schnellen Neutronen" to the German journal *Zeitschrift für Physik*.

After December 8 (Japan time), 1941, Japan became totally isolated from the world and the Tokyo cyclotron group became suddenly inactive for some reason, which I was not aware of.

It is interesting to note here that the above-mentioned manuscript submitted to the German journal one month after Japan's attack on Pearl Harbor, safely found its way to Berlin, even though World War II had already started in Europe in September 1939.

5. War-Time Research: 1941-1945

In September 1941, Professor Kimura hired a new technical assistant, whose name was Tamiya Asari, to cope with an increased number of samples for routine analyses, which were sent to his laboratories from various organizations. I was able to handle those request analyses in the early days, but it soon reached the point where I could no longer carry out my own work. I was therefore most happy to have Mr. Asari in our laboratories. While Mr. Ikawa was a quiet elderly gentleman, Mr. Asari was a young man of age 29 and he did not hesitate to talk to me and ask me numerous questions concerning chemical analysis. He was an extremely energetic young man and an excellent tennis player.

He told me that once his dream had been to become a Japanese tennis champion, but after meeting Professor Kimura he changed his mind and he hoped to someday become a great analytical chemist just like him. Early in the morning of his first day on the job, when I arrived at the Chemistry Building of the University, I was astonished to find Mr. Asari washing and cleaning with a piece of wet cloth and water, not only the surface of his laboratory bench and the inside and outside of all the drawers, but also the entire floor of the large laboratory, including the areas which were occupied by other people. I have never seen a laboratory assistant as eager as this young man from Akita Prefecture. I was extremely lucky, because his presence in our group enabled me to survive the most difficult period between December 8, 1941, and August 15, 1945, doing my doctor's thesis research.

It seemed to me that the government of Japan was not really interested in building an atomic bomb to win the war. They had more urgent problems at hand in all areas and one of them was a shortage of rare elements, such as lithium, cesium, and strontium. Professor Kimura first ordered Mr. Asari to measure the strontium contents of various rocks and minerals. This study soon led to the process of recovering strontium from the wastes of a barite factory located at the city of Fukuyama, in Okayama Prefecture. The

Imperial Japanese Navy was greatly impressed by this important war effort and awarded the Navy Secretary Prize jointly to Professor Kimura and Mr. Asari in April 1944. This was a great honor especially for Mr. Asari who had joined Professor Kimura's group only three years earlier as a laboratory assistant.

The next major war-time project was to extract lithium and cesium from the brines of Arima Hot Springs for the Japanese Imperial Navy. Arima, which is located near the famous port of Kobe, about 350 miles west of Tokyo, had been known as a spa, where rich people from Kobe and Osaka used to maintain their villas. The Japanese Imperial Navy was using this place as a rest and recreational area for the officers and sailors. Occasionally German submarines would enter the port of Kobe and their crews were also entertained at Arima.

A new graduate student named Hideo Yamatera was assigned to the Arima project by Professor Kimura at this time and Mr. Yamatera and I became room mates in Professor Kimura's old office in the Chemistry Building. Unlike Mr. Asari, he did not wash and clean the bench tops nor the entire floor, with a piece of wet cloth and water, but he was extremely polite and behaved exactly like a young samurai would have during the Tokugawa Period of Japan a century ago. Each time he entered the office in the morning and left the office in the evening, he bowed so deep to me that it was as if he came right out of the stage of the Kabuki theater as a young samurai wearing two Japanese swords in his belt, which made me feel that I had a young retainer who was ready to die for me in the battle field.

I later learned that Mr. Yamatera's ancestors were in fact high-ranking retainers of the Lord of Owari, who lived in the famous castle of Nagoya. The house in which he and his family live today is clearly marked on the map of Nagoya City, published during the Tokugawa Period - close to the castle of Nagoya.

The thesis problem which Professor Kimura assigned to Mr. Yamatera was "Extraction of cesium from Arima Hot Springs", which was, frankly speaking, not a type

of research to be considered worthy of publication in such journals as the *Journal of the Chemical Society of Japan*. As I recall, the academic record of Mr. Yamatera was outstanding, but he had the misfortune of having joined Professor Kimura's group in 1944 just at the time when the Empire of Japan was collapsing. I felt sorry for Mr. Yamatera and I attempted to encourage him by saying that we had a golden opportunity here to take advantage of the Arima project to extract a large amount of cesium from the water of the hot springs and look for the possible existence of element 87 in the sample by the use of the Siegbahn-type x-ray spectrograph which was not being used at that time in our laboratories. Element 87 (Francium) had actually been discovered and named "Actinium K" by M. Perey in Paris in 1939 as a member of the actinium series of the natural radioactive decay chain, but its half-life was only 22 minutes. I felt that there might exist an isotope of element 87, which was much more long-lived, and hence its natural abundance might be such that it would be detectable by the use of the x-ray spectrograph.

By November 1944, I managed to bring the Siegbahn x-ray spectrograph back in operation, which was not an easy task, because there were frequent power failures in Tokyo in those days and it was hard to maintain a steady voltage during the time of the experiments. The first x-ray spectrograph of milligram quantities of CsCl from Arima was taken on November 24, 1944. As I was operating the instrument in the second-floor laboratory, I heard loud sounds of air-raid sirens and the radio began announcing the arrival of large formations of B29's over the sky of Tokyo. This was the first of a series of full-scale bombing raids carried out by B29's from the island of Saipan, which would eventually destroy and burn most of the cities of Japan.

I was supposed to shut off the instrument immediately and go down to the bomb-shelter located in the basement of the Chemistry Building, but I decided to continue the operation because otherwise the precious sample would be wasted. Not only that, but if the bombs hit the Chemistry Department and destroyed the spectrograph, I would have had no opportunity to repeat the experiment. I heard big noises of falling bombs around the University campus, but none of them fell close to the Chemistry Building.

When I finally began developing the x-ray film in the dark room, I could not believe my eyes, because I saw a faint line beginning to appear at the center of the film where exactly the $L\alpha$ - x-ray line of element 87 should be located! Waves of B29's were still over the skies of Tokyo, but I was already thinking about writing a short manuscript on the discovery of element 87 to be submitted to the *Bulletin of the Chemical Society of Japan*. Since I was serving as one of the assistant editors of this journal published in the English, German or French languages, there would be no problem with the acceptance of this manuscript, but what if the building in which the journal was to be printed were destroyed, or if I died before peace returned to Japan, then there might be no way I could publish my important scientific discovery.

I soon realized however, that I did not have to worry about all these things, because I could not reproduce the experiment which I performed on that day in November 1944. The crude cesium chloride sample which I was using at first was further purified repeatedly, but none of the x-ray spectra taken thereafter showed any sign of the $L\alpha$ - line of element 87, and by the end of November 1944, I had just about given up the last hope of finding a new element by myself. The amounts of cesium chloride from Arima available for the experiments at that time were far less than one gram. If 100-gram or 1,000-gram quantities of CsC1 were available, one might be able to enrich *eka*-cesium by repeated fractional crystallization, and observe its presence in nature by the use of the x-ray spectrograph. I therefore asked Mr. Yamatera to take over the project and he replied to me that he would be delighted to do so.

At about this time in the fall of 1944, a military officer from the Eighth Imperial Army Research Institute visited me in my office and told me that his group was ready to launch a massive counter attack aimed directly at the mainland of the United States. Meteorologists in Japan knew that if a balloon with a timebomb were released in the upper atmosphere, it would travel eastward and reach the central part of the United States in 11.86 days if the average wind velocity was 10 meters per second, in 2.37 days if the average wind velocity was 50 meters per second, and in 1.19 days if the wind velocity was 100 meters per second. What the army officer wanted to know was, when the atom

bomb would become available so that he could place it on one of these balloons and drop it on the mainland of the United States!

My job at that time was to extract uranium from 600 tons of euxenite ore from Manchuria. Mr. Kunihiko Kigoshi, who was one of Professor Kimura's students working in Dr. Nishina's laboratories, was supposed to have separated ²³⁵U from the uranium extracted from the euxenite. Fig. 6 shows that a total of 13 balloon bombs actually did explode in the United States and Alaska during the period between January 7 and February 4, 1945. It is said that about 9,000 balloons were released, out of which a total of 287 bombs reached the targets (see Fig.6).

Mr. Yamatera kept the Siegbahn x-ray spectrograph in operation from the late fall of 1944 through the spring of 1945, when most of the city of Tokyo had been destroyed and the electricity and water supply had become essentially non-existent. He traveled back and forth between Arima and Tokyo, and eventually succeeded in isolating and purifying approximately 100 grams of CsC1. The x-ray spectrum taken with the purified sample of CsC1 showed a very faint line at the very position where the L α line of element 87 should have appeared. Both Mr. Yamatera and I agreed on this, and I went to Professor Kimura's office to show him the photograph. Professor Kimura looked at the faint spectral line and told me, smiling, that this was the K α line of bromine, not the L α line of element 87. The cesium sample apparently contained an extremely small amount of bromine as an impurity and we had been seeing the K α line of element 35, bromine. Professor Kimura had been taking x-ray spectra for twenty years, ever since he worked in Copenhagen under Professors Bohr and Hevesy in 1925. He was like an airplane pilot with an experience of 20 years of flying time, whereas Mr. Yamatera and I were like young inexperienced pilots with a combined flying time of less than one year.

The episode concerning the attempt made by Mr. Yamatera and me to discover element 87 in the water of Arima Hot Springs toward the end of World War II may appear simply as some sort of a tragicomedy, but it may provide us with a valuable lesson on how to decide on making a long-range plan for the future course of research in

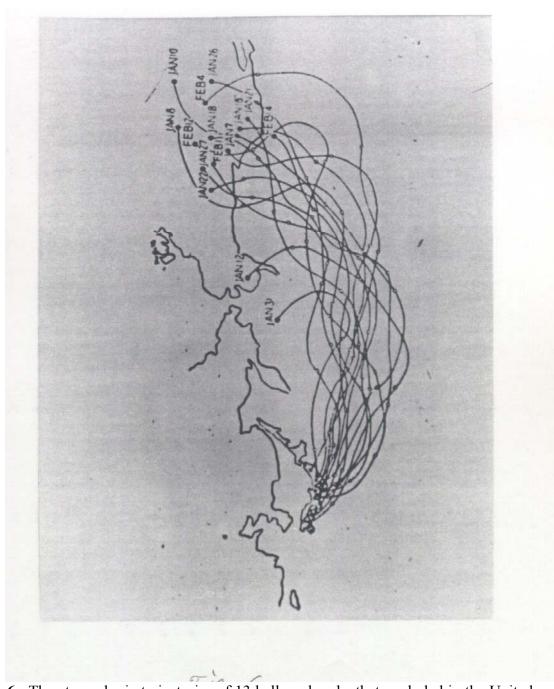


Figure 6. The atmospheric trajectories of 13 balloon bombs that exploded in the United States and Alaska during the period between January 7 and February 4, 1945.

science. It is generally accepted that the origin of geochemistry in Japan can be traced back to the year 1921 when the first report of chemical studies on minerals containing Rare Elements from East Asia by Yuji Shibata and Kenjiro Kimura appeared in the *Journal of the Chemical Society of Japan*, but nine years earlier in 1912, Professor Shibata went to Paris to study under Professor Georges Urbain, who in 1907 had discovered element 71, lutetium, which was the last of the rare earths to be discovered. In 1911, a year before the arrival of Shibata in Paris, Urbain thought he had isolated still another element, which he called "celtium" and which he believed fitted under zirconium in the periodic table.

Upon Professor Shibata's return to the Imperial University of Tokyo, the Adam-Hilger type E-2 quartz spectrograph made in England was introduced into the Chemistry Building and was in daily use in the 1930's when I worked there. Meanwhile, in Paris in 1914, Professor Urbain heard of Moseley's method of characterizing elements by the X-rays, so he traveled to Oxford to examine the rare earth sample, which he thought contained the new element "celtium". Moseley was able to prove however, that the sample contained nothing more than a mixture of known rare earths, and Professor Urbain is said to have been greatly impressed with all this. I imagine that Professor Shibata who was back in Tokyo by then, must also have been impressed. The missing element 72 located under zirconium in the periodic table was discovered by Coster and Hevesy in 1923 and named hafnium. They used the x-ray spectrograph to discover this new element.

In 1924, professor Kimura went to Copenhagen, Denmark, to study under Professor Hevesy and when he returned to Tokyo in 1926 the Siegbahn-type x-ray spectrograph, which was used by Mr. Yamatera and me in the fall of 1944 through the spring of 1945, was thus introduced into Professor Kimura's laboratories and brought into operation. All the rare minerals containing rare earth elements from Japan and a few Asian countries have been examined by the use of this equipment for a period of two decades, but none of the rare earths extracted from these minerals revealed the presence of x-ray lines of "missing" element 61. It so happened that the last of the stable new

elements discovered in the 20th century was element 75, rhenium, whose discovery was reported by Noddack *et al.* in 1925, when Professor Kimura was still in Copenhagen!

It is worthy of note here that when professor Shibata returned from Europe with the Adam-Hilger Type E-2 optical spectrograph, it was several years after the discovery of the last of the stable rare earth elements, lutetium, element 71, by Urbain in 1907. When professor Kimura returned from Europe with the Siegbahn-type x-ray spectrograph, it was also one year after the last of the stable elements to be discovered in natural minerals, namely element 75, rhenium, had been discovered by Noddack *et al.* in 1925. In each case, the gap between Europe and Japan seemed to become shorter and by 1936, when I entered the Imperial University of Tokyo, it appeared as if Japan had overtaken Europe as far as major scientific equipment was concerned, which is essential when being in competition with advanced nations in the world, even though this equipment became increasingly more expensive. In 1937, the Tokyo cyclotron was in operation, while no other European laboratories had access to this expensive and important equipment. Yet the most important discoveries during the 1930's were made by European scientists such as Segrè, Hahn and Strassmann, while others in Europe made certain tactical errors.

The Noddacks in Berlin, for example, thought in 1925 that they had discovered another new element, with an atomic number 43, which they named "masurium". They kept on trying to confirm its existence in nature, analyzing thousands of minerals and ore samples, which contained element 72, rhenium. As of 1949, x-ray spectral lines of "masurium" had been detected in 12 samples out of 24 ores and 32 concentrates and tailings from the rhenium production. The case of "masurium" was somewhat analogous to the attempt made by Mr. Yamatera and me to find element 87 in the cesium chloride samples extracted from Arima Hot Springs. Mr. Yamatera and I chose to search for element 87 in the water of Arima Hot Springs because of the facts that element 61 had always been absent in rare earth minerals analyzed in Professor Kimura's laboratories for a period of 20 years, from 1925 to 1945, and element 43 "masurium" had been reported to have been detected only a dozen times in Professor Noddack's laboratories in Berlin

during the same period. It seems to me that Professor Kimura's group in Tokyo and Professor Noddack's group in Berlin both made tactical errors in relying too heavily on the use of the x-ray spectrograph.

Four years before the discovery of fission by Hahn and Strassmann, Dr. Ida Noddack published an extremely important paper in *Angewandte Chemie*, volume **47**, page 653, <u>1934</u>, in which she pointed out the possibility that when the uranium atom is irradiated with slow neutrons, it may break up into more than one fairly large fragments. What she had envisioned in 1934 was the process of nuclear fission, but none of the scientists paid attention to her brilliant idea. She should have gone ahead at that time to prove her hypothesis by doing simple radiochemistry experiments which did not require expensive equipment like the x-ray spectrograph. If she had done so, she might have been able to discover the process of fission much earlier than Hahn and Strassmann and she could have won the Nobel Prize. Unfortunately however, she kept on looking for the missing element "masurium" by using the x-ray spectrograph for many years until her death in the 1970's.

In her letter dated 30 June 1972, addressed to me, Dr. Noddack bitterly complained about the fact that Professor Otto Hahn failed to properly reference her 1934 paper when he and Professor Strassmann discovered fission in 1939. Dr. Ida Noddack was born on February 25, 1896, in Wesel, Rhenish Prussia, three months earlier than Professor Kimura. The name rhenium comes from the Rhine River on whose shores she was born.

It seems to me that gross injustices have been done to both Dr. Ida Noddack and Professor Kimura by Professor Hahn, when he received the 1944 Nobel Prize in Chemistry. In his Nobel Lecture entitled "From the natural transmutations of uranium to its artificial fission", dated December 13, 1946, Hahn stated:

"--- We should point out here that other possibilities did not occur to anyone at that time. Since the discovery of the neutron and the application of artificial sources of radiation, a

large number of most unusual nuclear reactions had been discovered: the products were always either isotopes of the irradiated substances, or their next, or at most next-but-one, neighbours in the Periodic System; the possibility of a breakdown of heavy atomic nuclei into various light ones was considered as completely excluded."

I feel that it was a great insult to Dr. Ida Noddack not to mention her name here and reference her 1934 paper in making the above statement.

In regard to the important contributions made by the Nishina-Kimura group during the crucial period from 1937 to 1942, Hahn described them in only one sentence: "Japanese investigators found that, when fast neutrons were used, the fission of uranium proceeded more symmetrically than with slow ones." It was incredible that he mentioned not a single Japanese name, such as Nishina and Kimura, nor did he reference a single paper published by the Japanese scientists in scientific journals such as Nature, Physical Review or Zeitschrift für Physik in his 13 December 1946 Nobel Lecture. It was as if Germany and Japan had been enemies during World War II.

One reason for this may have been that, both Dr. Nishina and Professor Kimura studied in Copenhagen before the war under Professor Bohr, who was born in Denmark, and Professor Hevesy, who was born in Hungary. Hence there was no question that they were all enemy scientists from the standpoint of German scientists. Professors Fermi and Segrè were born in Italy, but they were playing crucial roles in the war effort of the United States and hence they were considered enemy scientists also. Nishina and Kimura were students of Bohr and Hevesy and hence they were likely to be faithful to their teachers. Moreover, Nishina and Kimura had been assisted in building the cyclotron by American scientists at the University of California, Berkeley, before World War II. On the other hand, Professor Hahn was not exactly popular among some German scientists because of his close ties to the brilliant Jewish lady physicist, Professor Lise Meitner, whose narrow escape from Germany in 1939 was made possible by the great help given to her by Professor Hahn.

December 13th, 1946, may not have been the happiest day of his life for Professor Hahn, judging from the last paragraph of his Nobel Lecture which reads as follows: "--- Both uranium 235 and plutonium are made in the United States. The result was the bombing of Hiroshima and Nagasaki."

It is perhaps important to note here that the key idea which enabled Professor Hahn to discover nuclear fission was the concept of radioactive indicator originated by none other than Professor Hevesy, who was the teacher of Professor Kimura. In his 13 December 1946 Nobel Lecture, Hahn made the following statement: "With the help of the <u>indicator method</u> we were able to prove without doubt that the 13-minute element of Fermi was neither a protactinium isotope, nor a uranium, actinium, or thorium." Our lack of experience in the radioactive indicator method at the Imperial University of Tokyo in 1939 was the direct cause for the failure of the Tokyo cyclotron group.

It is quite a coincidence that the 1943 Nobel Prize in Chemistry was awarded to Professor Hevesy "for his work on the use of isotopes as tracers in the study of chemical processes." The 1943 Nobel Prize was awarded to him on December 12, 1944, in Stockholm. It was about the time I asked Mr. Yamatera to take over the Arima project to isolate cesium chloride and look for element 87.

In his 12 December 1944 Nobel Lecture, Professor Hevesy stated: "The method of isotopic indicators had its ultimate origin in the Institute of Physics at the University of Manchester, which then was under the inspiring leadership of that great physicist, the late Lord (then Professor) Ernest Rutherford.

The cradle of radium is the Czecho-Slovakian town of Joachimstal; it was from Joachimstal pitchblende ore that Professor and Madame Curie isolated that element. The Austrian Government, the owner of these mines, generously supplied Professor Rutherford not only with radium, but also with the byproducts of radium production, equally important for the worker in the field of radioactivity. One of the most significant byproducts is radium D, which has a half-life period of 20 years and is found associated with the very substantial amounts of lead present in pitchblende. The Austrian

Government presented to Professor Rutherford several hundred kilograms of such <u>radiolead</u>. In view of its association with very large amounts of lead, which absorb the radiation emitted by radium D, this precious radioactive material nevertheless proved to be almost useless. When I met Professor Rutherford one day in 1911 in the basement of the laboratory where the <u>radio-lead</u> was stored, he addressed me in his friendly and informal way, saying "My boy, if you are worth your salt, you try to separate radium D from all that lead!" In those days, I was an enthusiastic young man and, on immediately starting to attack the problem suggested to me, I felt quite convinced that I would succeed. However, although I made numerous attempts to separate radium D from lead and worked for almost two years at this task I failed completely. In order to make the best of this depressing situation, I decided to use radium D as an indicator of lead, thus profiting from the inseparability of radium D from lead."

George de Hevesy was 28 years old in 1913, when he came up with the Nobel Prize-winning idea out of the seemingly hopeless research problem given to him by Ernest Rutherford. I was a 28-year old in 1945, when Mr. Yamatera and I worked on the Arima project and came out of it empty-handed. I did learn a lesson after all the work done in Arima and that was that the days of the x-ray spectrograph had long been over. In 1934, Professor Hevesy returned to Copenhagen and demonstrated the formation of new artificial radioactive isotopes and subsequently introduced a method of activation analysis based on neutron bombardment of the element to be investigated. This method was soon to replace the x-ray spectrographic method of chemical analysis.

It is interesting to speculate on what might have happened if Professor Kimura had spent two years studying under Professor Hevesy in Europe in the 1920's. Professor Kimura arrived at Copenhagen in March 1925, but Professor Hevesy left Copenhagen in July 1925 to become Professor of Physical Chemistry at Freiburg, Germany, while Professor Kimura decided to stay in Copenhagen. He therefore actually studied under Professor Hevesy for only three months and ended up spending the remaining twenty-one months in Copenhagen studying under two physicists, Niels Bohr and Yoshio Nishina, doing research with the x-ray spectrograph, which ten years later was destined to become

an obsolete instrument, when in 1937 the Tokyo cyclotron was brought into operation just two years before the discovery of fission by Hahn and Strassmann.

When I joined Professor Kimura's group in the spring of 1938, his laboratories were the best equipped in the world. In addition to the Adam-Hilger E-2 type quartz spectrograph and the Siegbahn-type x-ray spectrograph imported from Europe by Professors Shibata and Kimura during the 1910's and the 1920's, respectively, a polarograph was soon to become available from a company located in Kyoto, and I became in charge of setting it up in our laboratory. Upon the retirement of Professor Shibata from the Imperial University of Tokyo, Professor Kimura became Professor of Inorganic Chemistry and Professor Eiichi Minami succeeded him as Professor of Analytical Chemistry. Professor Minami had returned to Japan from Europe a few years earlier after doing research under Professor V. M. Goldschmidt in Göttingen, Germany and the Goldschmidt-type Optical Spectrograph, which was far more sensitive than the old Adam-Hilger E-2 type spectrograph, was to become operational. Moreover, we were to have access to Dr. Nishina's 26-inch cyclotron, which was the only cyclotron in operation anywhere in the world, except for the 37-inch cyclotron located in Berkeley, California.

Mr. Sakae Shinoda, who taught me how to operate the Siegbahn-type x-ray spectrograph when I was a new graduate student once remarked to me: "Professor Kimura loves to purchase new equipment one after another and he keeps us busy in maintaining them in good operating condition." Mr. Shinoda was the first student of Professor Kimura and served as his senior assistant ever since 1930. He was feared by many new students who took the first-year quantitative analysis laboratory course under his daily supervision. I felt like I was learning the ancient Japanese art of tea ceremony from a strict and highly disciplined master. My initial ambition when I was a new student was to someday become a senior assistant just like Mr. Shinoda. He soon became Dr. Shinoda and left Tokyo to become Professor of Analytical Chemistry at the Imperial University of Kyushu, on the western island of Kyushu where I was born. The quality of education in the first-year quantitative analysis laboratory course at the Imperial

University of Tokyo deteriorated rapidly soon after I replaced Mr. Shinoda as a senior assistant to Professor Kimura.

Our group at the Imperial University of Tokyo in the late-1930's was just like the Imperial Navy of Japan led by Admiral Isoroku Yamamoto, with the most powerful battleships in the world, with the best airplanes and carriers, but lacking a vital equipment called the radar. What was missing in our group was the vital concept of radioactive indicator, which had its origin in the mind of Professor George Hevesy before World War II. The concept of radioactive indicator was so beautifully utilized by Emilio Segrè in 1937 in his discovery of the first artificial element 43, technetium, and by Hahn and Strassmann in 1937 in their discovery of nuclear fission. The lack of experience in the use of radioactive indicator might not have existed in our group in the late 1930's, if Professor Kimura had followed Professor Hevesy to Freiburg in 1925 and studied under the inventor of the concept of radioactive indicator for two years until his return to Japan in 1927.

It seems to me, however, whether or not Professor Kimura stayed in Copenhagen or moved to Freiburg in 1925 should not have made any difference. Let us imagine that there had existed an active research program in Professor Kimura's laboratory on the study of radioactive indicator when I became his student, and if I were assigned to this program, the outcome would have been the same. It would have never occurred in my mind to try to use ²²⁸Ra (MsTh1) as a radioactive tracer to make sure that one of the radioactive isotopes present in Mr. Ikawa's samples was an isotope of barium and not radium. The problem simply lay in the fact that 99 out of 100 graduate students are neither smart enough nor truly motivated to become good scientists. I clearly belonged to the group of 99 out of 100 graduate students.

After the early spring of 1945, cities in Japan became the most dangerous place to live in the whole world, where tens of thousands of people were routinely killed overnight by the U.S. airplanes based on the islands in the South Pacific. Professor Kimura's laboratories were dispersed in several small cities and towns where it was

considered as relatively safe. Mr. Yamatera and I more or less alternated between Arima and Tokyo, making perilous journeys often in freight trains, which were often machine-gunned by thousands of American airplanes, which flew over Japan at will from the base on the island of Iwojima every day.

On Wednesday, May 30, 1945, I received two telegrams from Mr. Yamatera, who was in Tokyo at that time, while I was away at Arima. One said: "HOUSE DESTROYED IF YOU ARE UNABLE TO COME GIVE INSTRUCTIONS ON WHAT TO DO YAMATERA" and the other: "IF YOU MAKE IT TO TOKYO COME TO THE CHEMISTRY BUILDING YAMATERA". At this late stage of the war, telephone service was virtually nonexistent and train services were totally unreliable. Four days later on Sunday, June 3, 1945, a postcard describing more details arrived from Mr. Yamatera and at noon of that day I was able to leave Arima for Tokyo. Before the house was destroyed, Mr. Yamatera and his classmates had helped me in moving most of the books from my house to the Chemistry Building, so that my notes of Professor Kimura's lectures in Analytical Chemistry bound in the form of a book entitled "Kimura: Lehrbuch der Analytischen Chemie" has survived the war miraculously until today.

After living in Tokyo as a homeless person for one month sleeping in my office in the Chemistry Building, I returned to Arima on July 5th, 1945, with several pounds of rice and other forbidden items, which I had obtained through a black market merchant. On the way to Arima at the Osaka station, I was stopped by a policeman, who inspected the contents of my backpack. When I showed him the documents describing my official position as an Associate Professor of the Imperial University of Tokyo and the official war-time travel permit issued by the government to persons engaged in essential defense research, he told me that he was sorry to find an important person like me being involved in black marketing, but he would let me go free because he understood that I would not be able to perform my research without food. Among the forbidden items which I was carrying with me on that day were 700 seedlings of sweet potatoes, which I thought were needed to survive the summer of 1945. Mr. Yamatera assisted me in planting these seedlings in Arima. On Sunday, July 15, 1945, we had not yet finished planting the

seedlings and we were unaware of the fact that several thousand miles to the east across the Pacific Ocean, American scientists led by Professor Robert Oppenheimer were getting ready to test the first atom bomb at Alamogordo, New Mexico.

In the evening of Sunday, July 15, 1945, Enrico Fermi is said to have suddenly offered to take wagers from his fellow scientists on whether or not the bomb would ignite the atmosphere, and if so, whether it would merely destroy New Mexico or destroy the world. Fermi was obviously concerned about what Aston had said in his 1922 Nobel Lecture that: "--- In this event the whole of the hydrogen on the earth might be transformed at once and the success of the experiment published at large to the universe as a new star." Robert Oppenheimer is said to have assigned Edward Teller the task of trying to think of any imaginable trick or turn by which the explosion might escape its apparent bounds.

At 0529:45 (Mountain Standard Time) in the morning of July 16, 1945, the firing circuit of the bomb was closed. The test was a success and Emilio Segrè at the base camp recalled his first impression as follows: "--- We saw the whole sky flash with unbelievable brightness in spite of the very dark glasses we wore --- I believe that for a moment I thought the explosion might set fire to the atmosphere and thus finish the earth, even though I knew that this was not possible." The test of the world's first atom bomb was a great success. The atom bombs could be used safely against the enemy, without endangering the lives of the people living in friendly nations.

At 0836 Pacific War Time in the morning of July 16, 1945, four hours after the test at Alamogordo, New Mexico, the cruiser Indianapolis sailed under the Golden Gate at San Francisco and out to sea with its cargo containing two atom bombs to be used against Japan. The Indianapolis unloaded the bombs at Tinian on July 26 and sailed on to Guam and then toward Leyte in the Philippines, unescorted. The submarine 1-58 of the Japanese Imperial Navy, commanded by Lieutenant Commander Mochitsura Hashimoto found the Indianapolis and fired six torpedoes. According to Richard Rhodes' book "The Making of the Atomic Bomb" (Simon and Schuster, New York, 1986, pages 694 to 696),

out of 1,196 sailors on board the ship, some 850 men escaped to the sea and after an 84-hour ordeal, more than 500 men are said to have died, their bodies feeding sharks or lost to the depths of the sea.

It was no time for me to be planting the seedlings of sweet potatoes in Arima. Mr. Yamatera and I should have been back in Tokyo, since the radioactive particles injected into the atmosphere at Alamogordo, New Mexico, in the morning of July 16, 1945, should have been rapidly moving eastward, passing over Europe and Asia and arriving at Japan at the end of July.

Fig. 7 shows the estimated positions of the radioactive cloud from the first nuclear weapon's test. It is based on the studies carried out in Professor Yasuo Miyake's laboratories at the Geochemical Laboratory of the Meteorological Research Institute during the 1950's and 1960's concerning the trajectories of the radioactive fallout particles from the U.S. nuclear explosions at the Nevada Test Site and the Chinese test explosions conducted at Lop Nor, China (see Y. Miyake, Y. Sugiura and Y. Katsuragi, Radioactive fallout at Asahikawa, Hokkaido, in April 1955, J. Meteorol. Soc. Japan 34, page 226, 1956, and P. K. Kuroda, Y. Miyake and J. Nemoto, Strontium isotopes: Global circulation after the Chinese nuclear explosion of 14 May 1965, Science 150, page 1289, 1965). Fig. 7 shows the air movement at the height of 5650 meters in the troposphere, which is known to give a representative overall average value of the tropospheric air movement. It is assumed here that the meteorological conditions during the month of July 1945 were the same as those two decades later during the period between May 14 and June 11, 1965, when the Chinese government conducted its second nuclear test at Lop Nor, Sinkiang Province, China. The numbers in the circles show dates and the numbers between circles show average wind velocities in meters per second.

Mr. Yamatera and I should have set up an air-sampling system on the roof of the Chemistry Building and examined the radioactivity of air samples, which should have had unusually high beta-activities emitted by the fission products. Such information could have been extremely valuable to President Shigeru Nambara of the Imperial

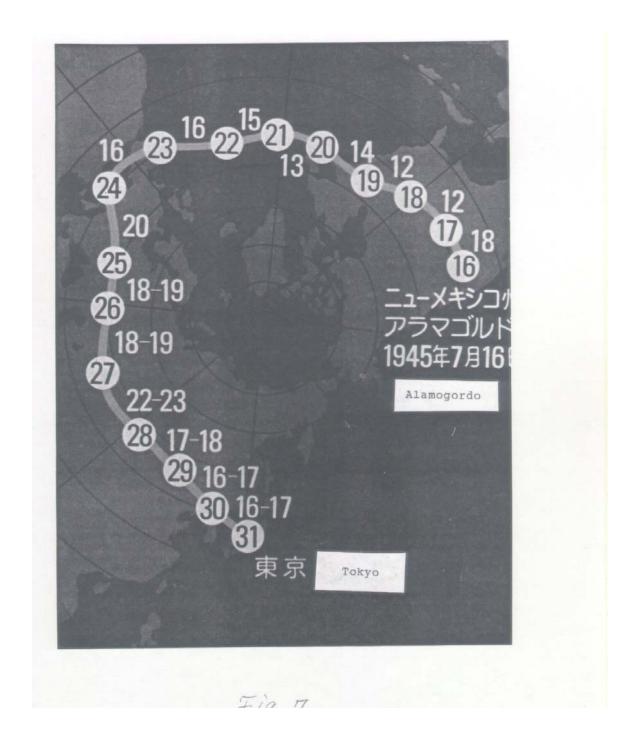


Figure 7. Estimated positions of the radioactive cloud from the first nuclear weapon's test at Alamogordo, New Mexico in the early morning of July 16, 1945.

University of Tokyo, who was at that time trying very hard to convince Prime Minister Kantaro Suzuki to accept the ultimatum issued to the government of Japan by the leaders of the allied nations in Potsdam, Germany, on July 27, 1945.

In the early morning of Monday, August 6, 1945, from 12:30 to 6:30 a.m., the Osaka and Kobe district was again attacked by B29's and flames of the burning cities were visible reaching high up into the sky beyond the dark silhouette of Rokko mountain. When the sun rose above the distant mountains, ashes from the cities were falling over Arima, while Rokko Mountain was covered with dense smoke. It did not appear to be an ideal day to travel to Tokyo, but I left Arima and made the journey which was relatively peaceful. I was not aware of the fact until I reached Tokyo, that a single bomb was dropped at 8:15 a.m. of that day over the city of Hiroshima, about 150 miles to the west of Kobe, and that 30,000 soldiers and some 28,000 to 30,000 laborers, as well as 75,821 civilians were killed instantly on the spot. The number of people killed in the following days and weeks by the effect of radiation is said to have increased the total number of persons killed to 210,000 to 240,000.

In the morning of August 7, 1945, I was summoned by Professor San-ichiro Mizushima to his office and I was asked my opinion concerning the nature of the bomb. It seemed that an official telephone call of inquiry concerning the bomb had reached Professor Mizushima's office from the government earlier in the morning and he was to return the call to express his opinion at once. He seemed to be hesitating to do so, however, because he was a physical chemist and was not familiar with problems related to nuclear weapons. Professor Kimura was gone, to Kanazawa, Ishikawa Prefecture, where one group of his staff members had established temporary laboratories a few months earlier. Professor Mizushima must have felt therefore that I may have been the only person who would be able to answer his question. I answered Professor Mizushima's question instantly, saying that I was convinced that the device was an atomic bomb. I was amazed at the speed with which American scientists had built the atom bomb, but there was no question in my mind that the world's brightest brains such as Fermi, Segrè and Seaborg were fully capable of accomplishing such a spectacular feat.

I felt that there was no time to be wasted and I immediately went to the Imperial Navy Headquarters near the palace to see a high-ranking officer in charge of naval research. When I found him in his office he was in a terrible mood, shouting at his subordinates, issuing new orders. He seemed to be pleased to see me, however, and he told me that he was no longer interested in trivial research, such as the Arima project. He then confided to me that he was going to initiate an atom bomb project of his own, so that I should return to the University at once, complete a literature survey on how to build an atom bomb and report back to him with a research proposal as soon as possible. I wanted to know if I was to be the principal investigator of the bomb project or was I supposed to write a research proposal for Professor Kimura, who was out of town and could not be reached easily, but I decided not to ask. At any rate, I now had a new assignment which appealed to me much more than the Arima project. In order to carry out this exciting new assignment, I should not be living a homeless person's life. On the way back to the University, I therefore stopped at the Gakushi-Kaikan (University Club) and reserved a single room, which was one of the very few decent hotel accommodations available in Tokyo at that time. This was a luxury which I could have never afforded under ordinary circumstances, but the war appeared to be ending soon and I could at least sleep comfortably and eat decent meals three times a day. The Japanese yen was soon to become almost useless anyway.

I spent the next eight days reading papers published during the 1930's by such scientists as Fermi, Segrè, Curie, Joliot and Seaborg in the libraries of the Imperial University of Tokyo. There were no scientific articles describing how to build an atom bomb, however, because American scientists had been keeping all their data strictly secret ever since the beginning of 1941, which was one year prior to the attack on Pearl Harbor. The war in Europe had started in September 1939 and there were few journals from Europe published after 1939. I did find a paper written in German, which I thought to be the most important. It was entitled "Kann der Energie-inhalt der Atomkerne technisch nutzbar gemacht werden?" (Can the energy content of the atomic nucleus be made useful?) and was written by a young theoretical physicist named Siegfried Flügge, who was then an assistant of Professor Lise Meitner. In this article Flügge pointed out

the possibility of a uranium chain reaction occurring in nature in uranium ore deposits such as those found in Bohemia and also in the state of Colorado in the United States. Flügge's idea appeared to be an extension of the idea which Aston had in mind in 1922, and I felt that if one could prove that such an event really took place in nature during the geological history of our planet earth, it would certainly revolutionize the basic concept of geochemistry. Apparently American scientists had already succeeded in gaining the fundamental knowledge required for the building of the atom bomb and the next logical step to be taken was to utilize this new knowledge and to attempt to prove that such an event had actually taken place during the geological history of the earth. I became extremely anxious to find out if this idea was being seriously pursued by American scientists in the summer of 1945.

The young German physicist named Siegfried Flügge and I were selected by Professor Segrè and Professor Seaborg nearly a half-century later to represent Germany and Japan, respectively, and to speak about the nuclear researches carried out in each country just before the second World War, at the International Symposium on "Fifty Years with Nuclear Fission", which was held in the large auditorium of the National Academy of Sciences in Washington, D.C. on April 26, 1989, to commemorate the anniversary of the discovery of fission. Dr. Flügge was unable to come to Washington because of his ill health and his paper was read by the symposium chairman. Because of the absence of Dr. Flügge, I became the only speaker representing a country which was on the losing side of World War II. It was one of the most difficult lectures which I have had to deliver in my lifetime. Among the slides I used in this lecture, one was the Japanese national flag and the other was a handsome photograph of Professor Kimura, which is shown at the beginning of this book. I took this picture in the summer of 1961 in front of the Tokyo office of the Institute of Atomic Energy Research, which was at that time located near Shinbashi station.

In the last paragraph of Dr. Flügge's speech entitled "How Fission Was Discovered", which was published in 1989 in volume 1, pages 26-29, of "50 Years with Nuclear Fission" by the American Nuclear Society, LaGrange Park, Illinois, U.S.A., he

made the following concluding remarks: "--- I remember a pleasant day in January, when we just had learned about the discovery (of fission by Hahn and Strassmann). On a walk after lunch we discussed in our small circle of young assistants why geological uranium deposits had never so far exploded by chain reaction. Back at the institute, I went to the library and, finding an old geological handbook, I looked it up to find out about uranium mines. You can still find the result of the inquiry in the final section of my survey article, written in May 1939 for "Naturwissenschaften" where I tried to discuss control of the chain reaction in order to avoid explosion. This publication, and a popular one that I published in a large daily newspaper two weeks before the outbreak of the war were meant to be a message to the community of science, the world over, and I am happy it was understood. After it, the curtain fell for years to come and for us there remained only the task to prevent the torch of science to be quenched in our country, however small its flames might become!"

It so happened that I picked up the concept of the Pre-Fermi reactor in the library of the Imperial University of Tokyo in the second week of August 1945.

The people who were staying at the Gakushi-Kaikan (University Club) were almost all middle-aged or older men, who had no place to live after their houses in Tokyo were destroyed and family members were sent to far-away villages. No one introduced himself to others, but we used to get together in the luxurious main sitting room with comfortable chairs every evening after supper. Everyone spoke in a low voice and with extreme caution, because there might be a plain-clothes policeman or an Army officer among us. All of them appeared to be graduates of the Imperial Universities and some worked at high government offices of Prime Minister Admiral Kantaro Suzuki's cabinet. I soon learned that high-level conferences were going on day and night and Prime Minister Suzuki was in frequent consultation with Emperor Hirohito. It appeared that the war would be ending within days. I only listened and never revealed my secret to others, namely, to try to build the bomb for the Imperial Navy, which by then had no battleships and no aircraft carriers left.

By Tuesday, August 14, 1945, I knew Japan would surrender unconditionally within twenty-four hours. Dr. Nishina at the Institute for Physico-Chemical Research had apparently ordered his men to destroy all the documents on the Army's uranium project. One of the assistants of Dr. Nishina came to my office in the Chemistry Building and gave me some of the documents, which he had been ordered to destroy. He told me that I would be the best person to preserve these documents for the future. I would be quite unlikely to be prosecuted as a war criminal, even if arrested by American soldiers, because my name appeared nowhere in these documents nor in published papers by the Nishina and Kimura group as a co-author. What he told me sounded quite reasonable and I promised him to keep these documents safely in my possession for the future. Forty-six years have elapsed since that day in August 1945 and I have kept my promise, but I have completely forgotten the name of the person to whom I made this promise and I should be most grateful to a reader of this book who might be able to identify that person.

I found most surprising statements in these documents. An Army document entitled "Concerning Uranium (U)" dated April 1943, describes that: "A lecturer at the Faculty of Science of the Imperial University of Kyoto, named Tokutaro Hagiwara, delivered a lecture entitled "Concerning super-explosive Uranium-235" at a meeting held by the Navy in May 1941, in which he stated that if an appreciable quantity of ²³⁵U could be manufactured and if a mixture of this ²³⁵U and an appropriate concentration of hydrogen with an appropriate volume could be prepared, ²³⁵U has the property of becoming a useful detonator substance."

The idea expressed by this young chemist named Tokutaro Hagiwara at the Imperial University of Kyoto in May 1941 was the basic principle of the hydrogen bomb. This means that Enrico Fermi and Edward Teller were not the first to conceive of using a nuclear chain reaction to initiate a thermonuclear reaction and this distinction belongs to the Japanese chemist Tokutaro Hagiwara of the Imperial University of Kyoto! Dr. Hagiwara also published in 1939 an article entitled "Liberation of neutrons in the nuclear explosion of uranium irradiated by thermal neutrons" in *The Review of Physical Chemistry of Japan* 13, page 145. The total number of neutrons emitted per fission was

2.6, according to the paper published by Hagiwara in 1939 and this value was in excellent agreement with that reported by American scientists W. H. Zinn and Leo Szilard, independently, in *The Physical Review* **56**, 619 (1939). By the way, Szilard was born in Budapest, Hungary, on February 11, 1898, and was a famous Hungarian-American physicist. He was born in the same city of the same country where George de Hevesy was born thirteen years earlier on August 1, 1885.

In 1934, Szilard and Chalmers published an important paper in the British journal *Nature*, volume **134**, page 462, which became well-known as the Szilard-Chalmers reaction. This was an extension of the concept of radioactive indicator put forward by Hevesy in 1913, and the concept of recoil effects of radioactive atoms was utilized by McMillan and Abelson in the experiments which led to the discovery of element 93, neptunium, in 1940.

Dr. Hagiwara had made these two major scientific contributions in 1939 and 1941, just prior to the Japanese attack on Pearl Harbor and yet these great scientific achievements remained unknown to the scientific community of Japan. The high-ranking navy officer whom I met a week before was apparently unaware of the fact as late as in 1945, that the Imperial Navy of Japan already had a key scientist with an idea and an experimental capability at least equal, if not greater than, those top scientists working on the atom bomb project in the United States. I wondered as to why the Imperial Navy of Japan had not appointed this young chemist to the top position in its bomb project. It was indeed ironic that I learned of Dr. Hagiwara's theory from the top secret documents of the Imperial Army of Japan which I obtained on August 14, 1945. It was apparent that there was no close cooperation between the Imperial Army and Navy of Japan, and they were acting like they were at war against each other, instead of against the United States.

Obviously there was no need for me to go back to see the naval officer at the Imperial Navy Headquarters after the Emperor's speech, which was broadcasted by the NHK radio at noon, Wednesday August 15, 1945. Japan had lost the war and I was alive. The weather was beautiful. I walked by the Imperial Palace, turned to the left instead of

going on in the direction of the Navy Department, and entered the Mitsukoshi department store, which to my surprise was open for business as usual. There was a sale of round straw hats, which was the kind popular among movie stars of Hollywood in the United States. I made my first purchase after World War II and paid one yen to buy the hat to replace my old military-style cap.

6. Research under the U.S. occupation: 1945-1949

I lived in Japan under the U.S. occupation for exactly four years from August 15, 1945 to August 15, 1949. These were the most humiliating years of my life. I was 28 in 1945 and I felt like a young baseball player, who watched a world series game from the dugout and witnessed our best team soundly being beaten by the opposing team, whose star players were world-famous scientists like Fermi, Segrè and Seaborg. I wanted to play in the next round of the world series game, but the best I could do in occupied Japan was to give up my career in baseball and change my job to that of someone who goes around the stadium selling beer and hot-dogs to the spectators, or else I could join a minor-league team in America and start my career all over again.

I was an Associate Professor of Chemistry of the Imperial University of Tokyo, the name of which was soon ordered by the Occupation Army to be changed to the University of Tokyo. Before August 15, 1945, salesmen from various companies which supplied us with reagents and glassware would make frequent visits to my office to take orders, and they delivered the items usually on the same day. After the unconditional surrender of Japan, I found myself behaving exactly the same way as those salesmen did. I made frequent visits to the Natural Resources Section of General MacArthur's GHQ to take orders for request analyses of rocks and minerals from various localities in Japan and from other Asian countries. Since I could make myself understood reasonably well, I soon became good friends with the Americans. I especially liked their habit of offering me a cigarette while I talked business with them and I soon found my business prospering. The request analyses were handled by Mr. Asari and his assistants and in the process, we could accumulate our inventories of reagents and glassware. I soon found out that the Americans were so rich that they did not care whether Mr. Asari needed one or ten bottles of fuming nitric acid for the analyses of hundreds of samples of ores and minerals.

The new 60-inch cyclotron just constructed by the group of Dr. Nishina together with the old cyclotron were dumped into Tokyo Bay by the U.S. Army soldiers, who thought they were machines to make the atom bomb. They did not destroy the Lauritsen-type electroscope, and with this left-over instrument I was able to maintain my research program on the radioactivities of Masutomi, Arima and Misasa Hot Springs for four more years, while Mr. Asari kept on doing his analysis of strontium in rocks and minerals.

When I sailed from Yokohama for San Francisco on board the S.S. General Gordon on August 12, 1949, Dr. Asari's doctoral dissertation entitled "Geochemical Distribution of Strontium" began appearing in the *Journal of the Chemical Society of Japan*.

7. The Incident in Chicago: 1957

In September 1957, Professor Kimura attended the first International Symposium on Radioisotopes held in Paris, France, as the chief Japanese delegate and he served also as Vice-President of the General Session. On this occasion, he received la Medaille de ville de Paris from the Mayor of Paris. On the way home from Paris, he visited Argonne National Laboratory which is located near Chicago, where I was doing research on the problem of stratospheric ⁹⁰Sr fallout. I served as his chauffeur while he stayed in the city of Chicago for a few days. I recall that the reception given to Professor Kimura by the Argonne National Laboratory was unusually cool and there were no lectures and welcome dinners planned for the visit of Professor Kimura. I waited at the airport for the arrival of his plane. I believe that two of his former students, Mr. Kunihiko Kigoshi and Mr. Tomitaro Ishimori, were with me at the airport, but no American scientists were there to welcome Professor Kimura. When Professor Kimura came out of the plane, accompanied by one of his assistants, Mr. Keiichiro Fuwa, his face was pale as he told me that he thought he might die when he noticed that one of the propellers of the airplane had stopped in flight.

I feel that the reason for the cool reception which Professor Kimura received in Chicago was two-fold. The first one was that at that time in the United States most scientists did not know who Kenjiro Kimura was. In 1946, Otto Hahn did not mention the name of Kimura anywhere in his Nobel Lecture, as I have mentioned earlier. When Glenn Seaborg received his 1951 Nobel Prize in Chemistry, he did reference the paper by Nishina, Yasaki, Ezoe, Kimura and Ikawa published in the *Physical Review*, Volume **57** (1940), page 1182, in the following manner:

"--- Before proceeding with a discussion of the other transuranium elements, I should like to say a few more words about neptunium. Another isotope, 237 Np, was discovered early in 1942 in collaboration with Wahl⁸⁾. This isotope is the decay product of the previous $1y^{9)$, $^{10)}$ known approximately 7-day beta particle emitting which is formed as the

result of an n,2n reaction on ^{238}U . The reactions for its production are shown in the next slide (Fig.6)".

$$238_{U} + n$$
 \longrightarrow $237_{U} + 2n$

$$\frac{237}{92}U \xrightarrow{\text{beta}} \frac{237}{6.6 \text{ days}} \text{ (2.20 x 10}^{6} \text{ years, alpha)}$$
Fig.6

The references cited by Seaborg are as follows:

- 8) A.C. Wahl and G.T. Seaborg, Nuclear properties of ²³⁷93, *Phys. Rev.* **73** (1948) 940 (report submitted April 14, 1942); NNES PPR, Vol. **14B**, *The Transuranium Elements : Research Papers*, Paper No. 1.5, McGraw-Hill Book Co., Inc., New York, 1949.
- 9) E.M. McMillan, Phys. Rev. 58 (1940) 178.
- 10) Y.Nishina, T.Yasaki. H. Ezoe, K. Kimura, and M. Ikawa, *Phys. Rev.* **57** (1940) 1182.

The average reader of the article may have felt that the discovery of 237 U was of minor importance compared with the discovery of 237 Np (Z = 93), which is true, but he may have concluded that it was discovered by McMillan before the group of Japanese investigators, because of the fact that the reference numbers are listed in reverse chronological order.

It seems to me that small, seemingly minor, inaccuracies which sometimes occur in the Nobel Lectures of the world's leading scientists, tend to have profound effects on the future course of science. A good example of this is the 8 December 1983 Nobel Lecture entitled "The Quest for the Origin of the Elements" by William A. Fowler

(*Science* **226**, 922, 1984), in which he conveniently forgets about mentioning the discovery of ²⁴⁴Pu in the early solar system, made in my laboratory at the University of Arkansas during the early 1960's. Instead, he refers to a review paper written by Wasserburg and Papanastassiou in 1982, most likely in an attempt to create an impression that the discovery of ²⁴⁴Pu in the early solar system was made at the California Institute of Technology. This seemingly minor inaccuracy which occurred in Professor Fowler's 1984 Nobel Lecture resulted in a state of total confusion which prevails until today, more than two decades after the successful Apollo 11 landing on the moon in 1969, and nearly destroyed a field of science called nuclear cosmochemistry.

The second reason was that many American scientists felt insulted by, instead of being grateful to, the scientists in Japan, who began a full-scale scientific research on radioactive fallout from a series of nuclear weapons tests being recklessly conducted in the South Pacific, notably on the island of Bikini, during the 1950's. When I joined the staff of Argonne National Laboratory in the summer of 1957, a local newspaper reported that news with a front page headline which said "JAP CHEMIST JOINS ARGONNE". That headline must have been about as large as that used when Japan attacked Pearl Harbor in 1941.

Another incident, besides the engine trouble of Professor Kimura's plane before landing at Chicago, occurred when I was taking Professor Kimura to his hotel late in the evening. My car collided with another car which was moving at a high speed on Lake Shore Drive along Lake Michigan. I immediately motioned the driver of the other car to pull out to a park, which was very dark. An Afro-American stepped out of his car and we both inspected the damages done to each other's cars.

My 4-year old 1953 Oldsmobile 88 with "rocket" engines, which was considered to be one of the luxury cars made in the U.S.A. after World War II, suffered only minor damage, while the other car was probably built in the 1930's and had numerous scars already before the collision, so that we were unable to identify the damages done by my car. I therefore asked him casually if he would mind forgetting about the whole thing,

because the passenger sitting in my car happened to be a very important person from Japan who was making an official visit to the great city of Chicago. The south side of Chicago was known to the world as being not exactly the safest place to live. It was somewhat like the black-market district west of the Ikebukuro station in Tokyo after World War II. People tended to be attacked, robbed and even killed routinely. I realized immediately after saying it, that I should not have told this man that Professor Kimura was a very important person from Japan. I should have settled the matter by myself, perhaps giving the man some money, not involving my great teacher.

What followed then was just like the scene from "Suzu-ga-mori" on the stage of the famous Kabuki Theatre, in which Banzui-in Chobei, played by Ichikawa-Danjuro is sitting alone in the abandoned "kago" (which is a basket palanquin hanging from a pole carried on the shoulders of two men). The stage is dark and there is no one there, except for, at a distance, Shirai-Gonpachi, a young samurai, who is inspecting his sword with which he has just killed several bandits. As if he was one of the bandits, the Afro-American approached my car, in which Professor Kimura was sitting alone. The only thing different was, that I was no Shirai-Gonpachi and I was unarmed. Professor Kimura did not utter a word and he just looked straight into the eyes of the Afro-American, who turned to me and said "O.K." with a smile. It was as if the Afro-American gentleman had seen the Kabuki play in Japan during the period of U.S. occupation and as if he knew the fate of the bandits who approached the "kago", in which Banzui-in Chobei was sitting alone. I hate to think of what might have happened if professor Kimura had been hurt in this incident and I feel very lucky it ended this way.

The research which I carried out at Argonne National Laboratory has remained unpublished in scientific journals up to the present time. The reason was that in the summer of 1958 I asked a well-known inorganic chemist at the Argonne National Laboratory, whom I thought of as my friend, to read the manuscript and to give me his impression of it. He kept the manuscript for a while and returned it to me saying that he thought it was an extremely interesting paper and he would be happy to publish it in a scientific journal if I would agree to add his name as a joint author. Instead of yielding to

his political pressure, I refused to publish the manuscript and promptly resigned from the Argonne National Laboratory to return to the University of Arkansas at Fayetteville, where the annual salary was about half of that paid at Argonne.

The research I did at Argonne laid the foundation of my work for the next thirty-three years. It was an extension of the concept of radioactive indicator put forward by George de Hevesy in 1913. If a series of rain samples are analyzed for ⁹⁰Sr, one obtains information on how the concentrations of ⁹⁰Sr in rainwater display a seasonal variation, increasing in the spring and decreasing in the fall. If the rain samples are analyzed also for ⁸⁹Sr and ¹⁴⁰Ba in addition to ⁹⁰Sr, one obtains additional information concerning the age of the radioactive isotopes found in the rainwater and hence the origin of the nuclear debris injected into the atmosphere.

In an article entitled "Nuclear Fission in the Early History of the Earth" published two years later in the British journal *Nature* **187**, 36 (1960), I have pointed out that the extinct ²⁴⁴Pu radionuclide with a half-life of 82 million years should have existed in nature during the early history of the solar system. More than three decades later, today I am busily engaged in a study concerning the ²⁴⁴Pu dating method for the early solar system, in which the relative numbers of atoms of ²³⁵U, ²³⁸U and ²⁴⁴Pu in meteorites and lunar samples during the early history of the solar system are calculated from the isotopic compositions of lead and xenon. The basic principle involved in the ²⁴⁴Pu dating method is essentially the same as that of the research on ⁹⁰Sr in rainwater, which I have carried out at Argonne in the 1957-1958 period, and both have their ultimate origin in the concept of radioactive indicator put forward by Professor Hevesy in 1913.

Results from these latest studies indicate that our current knowledge concerning the age of the earth and the solar system may be grossly in error and the key to the solution of this difficult problem may lie in some of the papers published by Professor Hevesy in the 1930's, and taught in great detail by Professor Kimura to me, for example, the paper concerning the determination of lead in rocks by the use of radioactive tracer,

which appears in my notes of Professor Kimura's lecture dated February 10, 1937 (See Fig.8).

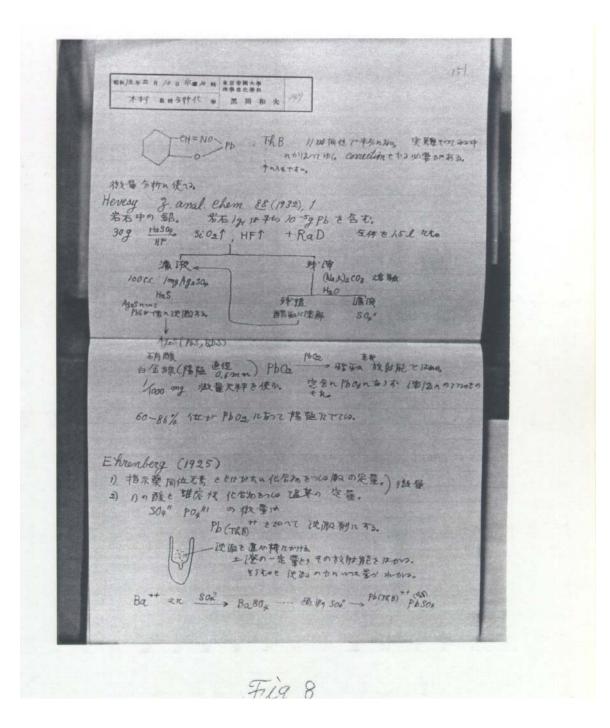


Figure 8. Errors in age estimates for the earth and the solar system may be solved by consideration of the papers published by Professor Hevesy in the 1930's on the determination of lead in rocks by the use of radioactive tracer, which appears in these notes of Professor Kimura's lecture dated February 10, 1937.

APPENDIX

Shown below are copies of Professor Paul K. Kuroda's original decay equations of radium and its decay products. The typed versions of these equations are shown at the bottom of page 24 and the top of page 25.

"Ra II"?
$$\frac{\beta}{\langle 1 \text{ Min.} \rangle}$$
 Ac I $\frac{\beta}{\langle 30 \text{ Min.} \rangle}$ Th?

"Ra II"? $\frac{\beta}{14\pm 2 \text{ Min.}}$ Ac II $\frac{\beta}{\sim 2.5 \text{ Hours}}$ Th?

"Ra III"? $\frac{\beta}{86\pm 6 \text{ Min.}}$ Ac III $\frac{\beta}{\text{many days}}$ Th?

"Ra IV"? $\frac{\beta}{250-300 \text{ Hours}}$ Ac IV $\frac{\beta}{\langle 40 \text{ Hours} \rangle}$ Th?

