Plutonium-244 in the early solar system and the Pre-Fermi natural reactor¹

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(Received December 2, 1991; Accepted February 17, 1992)

INTRODUCTION

I express my deep gratitude to the Geochemical Society of Japan for bestowing me the highest award, which carries the name of the late Professor Yuji Shibata of the Imperial University of Tokyo, the founding father of Geochemistry in Japan.

When I entered the University in 1936 and began attending a two-year course in Inorganic Chemistry taught by Professor Shibata, what impressed me most was that a large-scale research project on volcanoes in Japan, supported by the Imperial Academy of Science, was being maintained in his laboratories in the Chemistry Department, when it was a general custom in those days that chemistry professors seldom carried out researches based heavily on field work. I had the great fortune of being able to participate in Professor Shibata's research project as the youngest and most inexperienced member of his group for two years just before his retirement from the University, which coincided with the beginning of the war in the Pacific.

What I learned from Professor Shibata and his co-workers about geochemistry more than half a century ago gave me a strong incentive to undertake my studies on $^{244}$Pu in the early solar system and the natural reactors, many years later in the United States. It is therefore a great honor and pleasure for me to have this opportunity to speak to you on the current status of these ongoing investigations.

THE PRE-FERMI NATURAL REACTOR

I graduated from the Imperial University of Tokyo in the same year Hahn and Strassmann (1939) discovered fission. Soon thereafter, Professor Kenjiro Kimura, under whom I did my thesis work, told me to initiate a research work on the radioactivity of the water of Masutomi Radium Springs in Yamanashi Prefecture, jointly with another graduate student named Shinya Oana, who was doing his thesis work under the supervision of Professor Yuji Shibata on the density variations of natural waters associated with volcanoes. Our goal was to see if there was a certain relationship between the radioactivity and the heavy water concentrations of the water.

We found no clear relationship between them after studying the water of Masutomi Radium Springs for a period of about two years from 1939 through 1941, when the war began and our joint work was terminated (Oana and Kuroda, 1940, 1942). Although no definite conclusion

¹This paper is a condensed version of the acceptance speech delivered by the author on the occasion of his being awarded the Shibata Prize, at the Annual Meeting of the Geochemical Society of Japan, held in Matsuyama, Ehime Prefecture, Japan, on October 1, 1991.

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could be drawn from the early studies on the radioactive springs at Masutomi, I often wondered in those days whether or not a uranium ore deposit located deep under the ground might have been involved in some nuclear reactions in which the heavy water played an important role (for more details concerning the early history, see Kuroda, 1979, 1982, 1983, 1989, 1990a, b, 1991).

In 1952, I moved to the University of Arkansas at Fayetteville, Arkansas, and started my own research project, which was to measure the contents of various radioactive isotopes produced by the spontaneous fission of $^{238}$U in pitchblende ores (Kuroda and Edwards, 1954; Kuroda et al., 1956, 1957; Ashizawa and Kuroda, 1957; Parker and Kuroda, 1958; Heydeger and Kuroda, 1959; Kenna and Kuroda, 1960, 1961, 1964; Kuroda and Menon, 1961; Kuroda and Arino, 1964), with the ultimate objectives to establish the natural occurrences of element 43 (technetium) and element 61 (promethium), both of which had so far been regarded as "artificial" elements (Kenna and Kuroda, 1961, 1964; Attrep and Kuroda, 1968). The papers concerning the theory of natural reactors (Kuroda, 1956a, b) were by-products of the extensive radiochemical studies which were being carried out at the University of Arkansas during the 1950's.

In 1956, I predicted that a large uranium ore deposit could have become an operating pile during the geological history of the earth (Kuroda, 1956a, b). During the 1950's and 1960's, the idea of a critical uranium chain reaction occurring in nature was not taken seriously and it soon became almost completely forgotten, but sixteen years later, on September 25, 1972, the world’s scientific community learned of an extraordinary discovery made by research workers at the French Atomic Energy Establishment (Bodul et al., 1972; Neuilly et al., 1972; Baudin et al., 1972) that uranium had been found, in the deposit at Oklo in the Republic of Gabon, Africa, with an abnormal isotopic composition that led one to arrive at the conclusion that self-sustaining nuclear chain reactions had occurred in the remote past. The reaction site consisted of several bodies of very rich uranium ore, and more than 500 tons of uranium had been involved in the reactions with a quantity of energy released equal to about $100 \times 10^9$ kWh. The integrated neutron flux at certain points exceeded $1.5 \times 10^{21}$ neutrons per cm$^2$ (for more details, see "The Oklo Phenomenon", Proceedings of a Symposium, Libreville, 23-27 June 1975, published by the International Atomic Energy Agency, Vienna, 1975; excellent review articles published by Roth, 1977; and by Hagemann and Roth, 1978; a book written in the Japanese language by Fujii, 1985; and several review articles by Kuroda, 1975, 1982, 1983).

In 1976, Shukolyukov and co-workers (Shukolyukov et al., 1976; Shukolyukov and Min, 1977; Shukolyukov and Minh, 1977) reported that they found an anomalous fission xenon component in the Oklo reactors. The isotopic composition of this anomalous fission xenon is such that the abundances of $^{131}$Xe and $^{132}$Xe relative to $^{136}$Xe are markedly enhanced when compared to the relative fission yields from the thermal neutron-induced fission of $^{235}$U. The nature of this anomalous xenon from the Oklo reactor remained unexplained for many years, but Kuroda (1990a, b) pointed out that it can be attributed to the fact that $^{131}$Xe, $^{132}$Xe and $^{134}$Xe have fairly long-lived precursors: 8.04-day $^{131}$I, 78.2-hour $^{132}$Te and 42-minute $^{134}$Te, respectively.

Radionuclides which have been retained and preserved at the site of nuclear reactors have high melting and boiling points. Gaseous elements and elements with melting points lower than that of tellurium ($452^\circ$C) appear to have mostly migrated out of the reactor. About one percent of fission-produced xenon isotopes have been retained, however, and the isotopic compositions of small amounts of xenon released from the Oklo reactors were found to be abnormal in that the relative abundances of $^{131}$Xe and $^{132}$Xe were markedly enhanced.

The origin of this anomalous xenon can be attributed to a non-linear oscillatory mode of operation (Bilanovic and Harms, 1985) of the
reactors at temperatures of about 400°C, periodically being turned on and off, in a manner quite similar to the present-day geysers or intermittent hot springs. The time period during which the reactor was turned off was calculated to be about 3 hours from the observed ratios of $^{132}\text{Xe}$, $^{134}\text{Xe}$ and $^{136}\text{Xe}$ in the anomalous xenon (for more details, see Kuroda, 1990a, b; 1991).

**Plutonium-244 in Meteorites**

In 1957, the Soviet Union launched the Sputnik and newly elected President John F. Kennedy declared in 1960 that the United States would send Man to the moon before the decade was over. Reynolds (1960a, b) at Berkeley then made the important discovery that the xenon from the chondritic (stone) meteorite Richardton was heavily enriched in $^{129}\text{Xe}$. He concluded that this isotope almost certainly was formed from the radioactive decay of $^{129}\text{I}$ with a half-life of 16 million years, now extinct as a natural radioactivity but not so at the time of formation of the meteorite:

$$^{129}\text{I} \rightarrow ^{129}\text{Xe} \text{ (stable)}$$

Immediately after the discovery of $^{129}\text{I}$ in the early solar system, Kuroda (1960, 1961) pointed out that $^{244}\text{Pu}$ with a half-life of 82 million years should have also been present in the early solar system and the experimental evidence for its existence could be secured by searching for the presence in meteorites of excess heavy xenon.

![Diagram of the thorium series decay chain](image)

Fig. 1. The beginning of the thorium series decay chain.
isotopes $^{131}\text{Xe}$, $^{132}\text{Xe}$, $^{134}\text{Xe}$, and $^{135}\text{Xe}$ which are produced by the spontaneous fission of $^{244}\text{Pu}$ as shown in Fig. 1.

The presence of excess $^{244}\text{Pu}$ fission xenon was first reported by Rowe and Kuroda (1965) in the eucrite (pyroxene-plagioclase achondrite) Pasamonte and many supporting evidences for the existence of excess $^{244}\text{Pu}$ fission xenon in achondrites and ordinary chondrites have since been reported (see, for example, Rowe and Bogard, 1966; Merrihue, 1966; Kuroda et al., 1966; Hohenberg et al., 1967; Sabu and Kuroda, 1968; Meason and Rao, 1969; Wasserburg et al., 1969a, b; Reynolds et al., 1969). The final proof that the fission xenon observed in the meteorites was in fact the spontaneous fission decay product of $^{244}\text{Pu}$ was reported by Alexander et al. (1971). They used 13.0 mg of “pure” $^{244}\text{Pu}$ (as $^{244}\text{PuO}_2$) and measured the relative mass yields of the fissionogenic xenon isotopes with a mass spectrometer. Their measurements agreed almost perfectly with the $^{244}\text{Pu}$ spontaneous fission mass yields deduced from the meteorite data, as shown in Table 1.

### Plutonium-244 in Carbonaceous Chondrites

Six months before the report by Rowe and Kuroda (1965) appeared in the February 1, 1965 issue of the Journal of Geophysical Research, a paper entitled “Rare Gases in the Chondrite Renazzo” by Reynolds and Turner (1964) appeared in the August 1, 1964 issue of the same journal. In this paper, Reynolds and Turner (1964) reported that they discovered the presence of roughly $5 \times 10^{-12}$ (cc STP/g) of excess fission xenon ($^{136}\text{Xe}$) in the carbonaceous chondrite Renazzo and, although the amount of excess xenon found in Renazzo appeared to be greater than expected by at least a factor of 2, they noted that “...this discrepancy alone is not sufficient to rule against the possibility of $^{244}\text{Pu}$ fission gas in Renazzo.”

Three years later, Funk et al. (1967) re-examined the xenon isotope data for the carbonaceous chondrites Murray (Reynolds, 1963) and Renazzo (Reynolds and Turner, 1964) and reported that they confirmed the presence of exceptionally large amounts of fission xenon postulated by Reynolds’ group, as shown in Table 2. They reported to have demonstrated that three possible complications, namely, the presence of an atmospheric component, a cosmic-ray component and diffusion effects (mass-fractionation) were not capable of explaining away the large excesses of fission xenon observed and stated that: “We have concluded, with Reynolds et al., that the amounts of fission xenon in Renazzo and Murray are incompatible with the model of continuous galactic nucleosynthesis (Burbidge et al., 1957), if the fission xenon is from $^{244}\text{Pu}$ which decayed in situ.”

The presence of large excesses of fission xenon was soon reported in other carbonaceous chondrites, such as Mokoia (Rowe, 1968; Manuel et al., 1972a), Leoville (Manuel et al., 1970) and Allende (Manuel et al., 1972a) and attempts to explain the origin of this strange xenon component were made by many investigators. Manuel et al. (1972b), for example, argued that carbonaceous chondrites contained isotopically distinct components which could not be explained by the occurrences of nuclear or fractionation processes within these meteorites and suggested that the large excesses of fission xenon found in these meteorites may be the product of galactic nucleosynthesis, which had not been uniformly mixed with the solar system materials (see also Sabu et al., 1974; Manuel and Sabu, 1975; Sabu and Manuel, 1980).

Anders et al. (1975), on the other hand,
speculated that one of the unknown superheavy elements (element 115, 114, or 113) may have had an isotope with a half-life in the range of $10^7$ to $10^8$ years, which is too short to survive to the present day, but long enough to leave detectable effects in meteorites, and this isotope may have been present in the carbonaceous chondrites and decayed to $^{131-136}$Xe by spontaneous fission (see also Lewis et al., 1975).

Srinivasan and Anders (1978) reported, however, that the carbonaceous chondrite Murray chison contained a new type of xenon component highly enriched in five of nine stable isotopes, mass numbers 128 to 132, and these patterns were highly suggestive of the s-process nucleosynthesis believed to take place in red giants. It is also worthy of note that Lewis et al. (1983) reported that carbon and chromatite fractions from the Allende meteorite that contain isotopically anomalous xenon-131 to xenon-136 (carbonaceous chondrite fission or CCF xenon) at up to $5 \times 10^{11}$ atoms per gram, showed no detectable isotopic anomalies in barium-130 to barium-138, and this seemed to rule out the possibility that the CCF xenon was formed by in situ fission of an extinct superheavy element. They concluded therefore that the CCF xenon and its carbonaceous carrier were relics from stellar nucleosynthesis (see also Swart et al., 1983; Wieler et al., 1991).

Kuroda (1971) emphasized, however, that the xenon isotope data should be interpreted in terms of the alterations of the relative abundances of each of the nine stable isotopes of xenon caused by the processes of a) mass-fractionation, b) spallation and c) neutron-capture reactions (plus the decays of $^{129}$I and $^{244}$Pu to produce excesses of $^{129}$Xe and $^{131-136}$Xe, respectively) (see also, Kuroda et al., 1974, 1975; Kuroda, 1976a, b, 1982; Kuroda and Sheng, 1986a, b; Saebo and Kuroda, 1986a, b). It is important to note here that Funk et al. (1967) reported earlier that the processes such as those mentioned above seemed to play a relatively minor role in altering the isotopic composition of xenon found in carbonaceous chondrites, but a vast amount of experimental data which have been accumulated since 1967 seem to indicate that it may not necessarily be the case.

Figures 2 and 3 compare the 3-isotope plots of $^{134}$Xe/$^{136}$Xe vs $^{134}$Xe/$^{136}$Xe where $i=124$, 128, 129, 130, 131 and 132 for the isotopic compositions of xenon released from achondrites (Myers and Kuroda, 1991a) and carbonaceous chondrites (Myers and Kuroda, 1991c). The data points for a total of 213 analyses of xenon released from bulk samples and temperature fractions of 35 achondrites and a total of 80 analyses including xenon fractions released at different temperatures and the total release from bulk samples of the carbonaceous chondrites Renazzo, Mokoia and Groznaya are plotted in Fig. 2 and Fig. 3, respectively. The point $P$ in these figures is Takaoka (1972)'s primitive xenon, $A$ is the atmospheric xenon, $X$ is the so-called CCF xenon, and $F$ is the $^{244}$Pu fission xenon (Alexander et al., 1971). The curve PM is the mass-fractionation line for the atmospheric

Table 2. Estimation of the excess fission xenon ($^{134}$Xe) contents and the initial $^{244}$Pu/$^{238}$U ratios in the carbonaceous chondrites Murray and Renazzo (Funk et al., 1967)

<table>
<thead>
<tr>
<th>Meteorite</th>
<th>$^{134}$Xe (10$^{-11}$ cc STP/g)</th>
<th>($^{244}$Pu/$^{238}$U)$_b$ (atom/atom)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) Murray</td>
<td>$&gt;$52</td>
<td>$&gt;$0.26</td>
<td>Reynolds (1963) and Hohenberg et al. (1967)</td>
</tr>
<tr>
<td></td>
<td>$&gt;$90±40</td>
<td>$&gt;$0.45±0.17</td>
<td>Funk et al. (1967)</td>
</tr>
<tr>
<td>(2) Renazzo</td>
<td>40</td>
<td>0.19</td>
<td>Reynolds and Turner (1964)</td>
</tr>
<tr>
<td></td>
<td>15±6</td>
<td>0.08±0.02</td>
<td>Funk et al. (1967)</td>
</tr>
<tr>
<td>(3) Steady State Theory</td>
<td>—</td>
<td>$\leq 0.05$</td>
<td>Burbidge et al. (1957), Fowler (personal communication, 1966)</td>
</tr>
</tbody>
</table>
Fig. 2. Three isotope correlation plots for xenon released from achondrites (Myers and Kuroda, 1991c). Point A is the composition of atmospheric xenon, F is $^{244}$Pu fission xenon (Alexander et al., 1971), P is the primitive xenon of Takaoka (1972) and X is the CCF xenon (Myers and Kuroda, 1989). The curve PM is the mass fractionation line for atmospheric xenon.
\[ \frac{{^{128}Xe}}{{^{136}Xe}} \]

\[ \frac{{^{134}Xe}}{{^{136}Xe}} \]

\[ \frac{{^{129}Xe}}{{^{136}Xe}} \]

\[ \frac{{^{134}Xe}}{{^{136}Xe}} \]
xenon.

Figure 2 shows that many of the data points tend to lie along the lines connecting the points $P$ (primitive xenon) or $A$ (atmospheric xenon) and $F$ ($^{244}$Pu fission xenon) in the plot of $^{134}$Xe/$^{136}$Xe vs $^{134}$Xe/$^{136}$Xe ratios for $i=132$, demonstrating the fact that we are dealing here mostly with the mixtures of $^{244}$Pu fission xenon and a mass-fractionated primitive xenon $P$ and it is possible to calculate the approximate amounts of $^{244}$Pu fission xenon present in the samples from these data. In the cases of the plots of $^{132}$Xe/$^{136}$Xe vs $^{134}$Xe/$^{136}$Xe ratios for $i=124, 126$ and $128$, most of the data points lie far above the line $PF$, indicating that the relative abundances of $^{134}$Xe, $^{136}$Xe and $^{128}$Xe are severely altered by the processes of a) mass-fractionation, b) spallation and c) neutron-capture. In the cases for the plots for $i=129, 130$ and $131$, the scatter of the data points is less pronounced and, although many of them lie along the lines of $PF$ and $PM$, it is virtually impossible to estimate the amounts of $^{244}$Pu fission xenon in the samples from these xenon isotope data.

Figure 3 shows that a scatter of the data points is less pronounced as compared with the case of achondrites shown in Fig. 2 and the data points in the plots of $^{132}$Xe/$^{136}$Xe vs $^{134}$Xe/$^{136}$Xe ratios for $i=130, 131$ and $132$ tend to lie along the line $PX$ and this creates an impression that there exists a xenon component whose isotopic composition $X$ is different from that of the $^{244}$Pu fission xenon. It is to be noted here, however, that the lines $PX$ and $PM$ lie close to each other in these plots and there is a clear trend that the data points tend to deviate upward from the line $PM$ in the cases of the plots for $i=130$ and $131$, while the opposite appears to be the case for $i=132$, reflecting the fact that the relative abundances of $^{136}$Xe and $^{131}$Xe can be more easily altered than that of $^{132}$Xe by the above-mentioned processes a), b) and c). This means that the fact that the data points lie along the line $PX$ in these plots does not necessarily support the view that the carbonaceous chondrites contain excess fission xenon whose isotopic composition is different from the $^{244}$Pu fission xenon. It is more likely that we are dealing here with mixtures of $^{244}$Pu fission xenon and a mass-fractionated primitive xenon $P$, just as it was the case for the
Fig. 3. Three isotope correlation plots for xenon released from the carbonaceous chondrites Renazzo (○), Mokoia (+) and Groznaya (∆) (Myers and Kuroda, 1991a). Point A is the composition of atmospheric xenon, F is $^{244}$Pu fission xenon (Alexander et al., 1971), P is the primitive xenon of Takaoka (1972) and X is the CCF xenon (Myers and Kuroda, 1989). The curve PM is the mass fractionation line for atmospheric xenon.
$^{244}\text{Pu}$ in the early solar system and natural reactors

c)

\[ \frac{^{128}\text{Xe}}{^{136}\text{Xe}} \]

\[ \frac{^{134}\text{Xe}}{^{136}\text{Xe}} \]

d)

\[ \frac{^{129}\text{Xe}}{^{136}\text{Xe}} \]

\[ \frac{^{134}\text{Xe}}{^{136}\text{Xe}} \]
xenon found in chondrites and achondrites, the only difference being that the carbonaceous chondrites contain far greater amounts of trapped (mass-fractionated primitive) xenon relative to the $^{244}$Pu fission xenon than the xenon found in the chondrites and achondrites.

These results indicate that it is virtually impossible to determine the contents of $^{244}$Pu fission xenon in meteorites without taking into consideration the alterations of relative abundances of all the stable isotopes of xenon. We have therefore developed a new method in which a computer is used to calculate the isotopic compositions of trapped xenon and the amounts of $^{244}$Pu fission xenon found in each of the xenon fractions released from the meteorites (for details concerning the method of calculations see Kuroda, 1989; Kuroda and Myers, 1989, 1991a, b, c, d, e; Myers and Kuroda, 1991a, b, c, d).

Table 3 shows that the amounts of the $^{244}$Pu fission xenon thus calculated for six carbonaceous chondrites are all in the range of 22 to $34 \times 10^{-12}$ (cc STP $^{136}$Xe/g) and are intermediate between the values of $40 \times 10^{-12}$ (cc STP $^{134}$Xe/g) and $(15 \pm 6) \times 10^{-12}$ (cc STP $^{134}$Xe/g) reported by Reynolds and Turner (1964) and Funk et al. (1967), respectively, for the carbonaceous chondrite Renazzo (see Table 2). Initial ratios of plutonium to uranium ($^{244}$Pu/$^{238}$U)$_0$, were calculated from the contents of $^{244}$Pu fission xenon ($^{136}$Xe) and uranium. The $^{244}$Pu ages were then calculated as described by Kuroda (1989) using the value of ($^{244}$Pu/$^{238}$U)$_0$ = 0.0117 ± 0.0014 (atom/atom) for the Kapeota meteorite as a reference standard and accepting its age 4560 million years to be correct (Rowe, 1970). The values of ($^{244}$Pu/$^{238}$U)$_0$ thus calculated for six carbonaceous chondrites range from 0.114 to 0.170 (atom/atom) and they appear to have started to retain their xenon 4800 to 4900 million years ago. The values of ($^{244}$Pu/$^{238}$U)$_b$ for four chondrites range from 0.0070 to 0.0130 (atom/atom) and their $^{244}$Pu ages range from 4500 to 4573 million years. The values of ($^{244}$Pu/$^{238}$U)$_b$ for four achondrites range from 0.0018 to 0.0117 (atom/atom) and their $^{244}$Pu ages range from 4337 to 4560 million years.

The $^{244}$Pu ages thus obtained for the car-
bonaceous chondrites are much greater than the value of (4550±70) million years for the earth and meteorites reported by Patterson (1956) and also the values of (4511±42) and (4496±10) million years for the $^{207}\text{Pb}/^{206}\text{Pb}$ ages reported by Tatsumoto et al. (1973) for the Murray and Allende meteorites, respectively. It is extremely difficult, however, to determine the lead-isotope ages of bulk samples of carbonaceous chondrites because the isotopic compositions of lead found in this group of meteorites are quite similar to that of the primordial lead. Chen and Wassenburg (1981) reported that the isotopic compositions of lead found in acid-soluble phases of the Allende coarse-grained inclusions gave a mean $^{207}\text{Pb}/^{206}\text{Pb}$ model age of (4559±15) million years, while Droz et al. (1977) reported the values of $(^{244}\text{Pu}/^{238}\text{U})_0$ as high as $>0.14±0.02$ and $0.087±0.011$ (atom/atom) for two fine-grained inclusions of the Allende meteorite.

**Plutonium-244 in Lunar Samples**

In the first report on the examination of the samples brought to earth from the 20 July 1969 Apollo 11 landing on the moon, members of the Lunar Sample Preliminary Examination Team (LSPET, 1969) wrote that the isotopic compositions of xenon found in the fines and in a breccia resembled those of trapped xenon found in carbonaceous chondrites and that decay products of the extinct nuclides $^{129}\text{I}$ and $^{244}\text{Pu}$ appeared to be absent in lunar samples. These early results seemed to indicate that the moon was not quite as old as it had been thought earlier and the results from the age determinations of lunar
samples by the K–Ar and Rb–Sr dating methods seemed to support this view.

Soon after the lunar samples were returned from the 5 February 1971 Apollo 14 landing on the moon, the Lunar Sample Preliminary Examination Team (LSPET, 1971) reported that the lunar breccia 14301, one of the five samples whose isotopic compositions of xenon were determined mass-spectrometrically, contained as much as 150 \times 10^{-12} \text{ cc STP} \ ^{136}\text{Xe} per gram of fissionogenic xenon. The uranium content of 14301 was 3.5 ppm and the amount of excess fissionogenic xenon which they found in this sample was clearly much more than could have been produced by the spontaneous fission of \(^{238}\text{U}\) in 4.5 billion years. They reported, however, that there appeared to be no need to invoke extinct radionuclides to explain the observed abundances of \(^{129}\text{Xe}\) and \(^{136}\text{Xe}\), with the possible exception of sample 14301.

The case of sample 14301 soon ceased to be an exception and the presence of \(^{244}\text{Pu}\) fission xenon in lunar samples was reported by several investigators (see, for example, Drozd et al., 1972; Behrmann et al., 1973; Drozd et al., 1975, 1976; Swindle et al., 1985). These investigators have concluded that the excesses of \(^{244}\text{Pu}\) fission xenon were not due to \textit{in situ} decay of \(^{244}\text{Pu}\), but they were "parentless". Which means that the daughter products were incorporated onto grain surfaces following decay of the parent nuclide elsewhere (Swindle et al., 1985).

Table 4 shows the contents of uranium, \(^{244}\text{Pu}\) fission xenon, and the values of \((^{244}\text{Pu}/^{238}\text{U})_0\) and \(^{244}\text{Pu}\) ages of a total of 11 samples of fines, regolith, breccias and rock (Kuroda and Myers, 1971c). These values were calculated in the same manner as in the cases of meteorites shown in Table 3. Uranium contents of lunar samples are generally much higher than those of the meteorites and, accordingly, the amounts of \(^{244}\text{Pu}\) fission xenon are also considerably higher.

Table 4. Plutonium-244 fission xenon in lunar samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>(\text{U (ppm)})</th>
<th>(^{136}\text{Xe} (10^{-12} \text{ cc STP/g}))</th>
<th>((^{244}\text{Pu}/^{238}\text{U})_0) (atom/atom)</th>
<th>(^{244}\text{Pu}) age (million years)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>(A) Lunar Fines and Regolith</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1608-3-26</td>
<td>0.25</td>
<td>738 ± 155</td>
<td>0.206 ± 0.040</td>
<td>4904 ± 28</td>
</tr>
<tr>
<td>10084-12</td>
<td>0.46 ± 0.10</td>
<td>602</td>
<td>0.091 ± 0.020</td>
<td>4808 ± 30</td>
</tr>
<tr>
<td>10084-59</td>
<td>0.46 ± 0.10</td>
<td>336</td>
<td>0.051 ± 0.011</td>
<td>4738 ± 30</td>
</tr>
<tr>
<td>10084-29</td>
<td>0.46 ± 0.10</td>
<td>141</td>
<td>0.021 ± 0.005</td>
<td>4631 ± 25</td>
</tr>
<tr>
<td><strong>(B) Lunar Breccias</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14313</td>
<td>3.23</td>
<td>1019 ± 200</td>
<td>0.022 ± 0.004</td>
<td>4636 ± 26</td>
</tr>
<tr>
<td>67455</td>
<td>0.05</td>
<td>6.7 ± 1.0</td>
<td>0.0083 ± 0.0014</td>
<td>4533 ± 19</td>
</tr>
<tr>
<td>14055, 3</td>
<td>3.68</td>
<td>476 ± 46</td>
<td>0.0090 ± 0.0009</td>
<td>4529 ± 12</td>
</tr>
<tr>
<td>14047</td>
<td>3.39</td>
<td>393 ± 41</td>
<td>0.0081 ± 0.0008</td>
<td>4516 ± 13</td>
</tr>
<tr>
<td>14301</td>
<td>3.63</td>
<td>282 ± 13</td>
<td>0.0054 ± 0.0002</td>
<td>4468 ± 5</td>
</tr>
<tr>
<td>14318</td>
<td>4.0</td>
<td>236 ± 11</td>
<td>0.0041 ± 0.0002</td>
<td>4435 ± 5</td>
</tr>
<tr>
<td><strong>(C) Lunar Rocks</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10057</td>
<td>0.78 ± 0.16</td>
<td>8.1*</td>
<td>0.00051 ± 0.00014</td>
<td>4181 ± 30</td>
</tr>
</tbody>
</table>

* This includes about 2.3 \times 10^{-12} (cc STP/g) of \(^{136}\text{Xe}\) from the spontaneous fission of \(^{238}\text{U}\).
than those of the meteorites. The values of \(^{238}\text{U}/^{235}\text{U}\) for lunar fines and regolith range from 0.021 to 0.206 (atom/atom) and their \(^{244}\text{Pu}\) ages from 4631 to 4904 million years. The values of \(^{238}\text{U}/^{235}\text{U}\) for six samples of lunar breccias range from 0.0041 to 0.022 (atom/atom) and their \(^{244}\text{Pu}\) ages from 4435 to 4636 million years. The lunar rock 10057 contains \(8.1 \times 10^{-12}\) (cc STP \(^{136}\text{Xe}/\text{g}\)) of excess fission xenon, which includes about \(2.3 \times 10^{-12}\) (cc STP \(^{136}\text{Xe}/\text{g}\)) of fission xenon from the spontaneous fission of \(^{238}\text{U}\) (Kuroda and Myers, 1991e) and its \(^{244}\text{Pu}\) age is 4181 million years.

The \(^{244}\text{Pu}\) ages of lunar samples thus calculated are in general agreement with their lead-isotope ages. For example, according to Tatsumoto and Rosholt (1970), the \(^{206}\text{Pb}/^{238}\text{U}, ^{207}\text{Pb}/^{235}\text{U}\) and \(^{207}\text{Pb}/^{206}\text{Pb}\) ages of lunar rock 10057 are 4089, 4146 and 4173 million years, respectively, in excellent agreement with the \(^{244}\text{Pu}\) age of 4181 million years. They also reported the \(^{206}\text{Pb}/^{238}\text{U}, ^{207}\text{Pb}/^{235}\text{U}\) and \(^{207}\text{Pb}/^{206}\text{Pb}\) ages of lunar fine 10084 to be 4685, 4668 and 4659 million years, while the \(^{244}\text{Pu}\) age of 10084 ranges from 4631 to 4808 million years as shown in Table 4. According to Tatsumoto (1973), the \(^{206}\text{Pb}/^{238}\text{U}, ^{207}\text{Pb}/^{235}\text{U}, ^{207}\text{Pb}/^{206}\text{Pb}\) and \(^{208}\text{Pb}/^{232}\text{Th}\) ages of Luna 20 soil are 5130, 5020, 4980 and 4950 million years, respectively. Although the xenon isotope data for the Luna 20 soil sample are not available, the \(^{244}\text{Pu}\) age of the Luna 16 regolith 1608–3–26 is 4904 million years, as shown in Table 4. It is also interesting to recall that Silver (1970a, b) pointed out that the leads found in the Apollo 11 fines and breccia are mixtures of extraordinarily heterogeneous components having \(^{207}\text{Pb}/^{206}\text{Pb}\) ratios which vary by more than 50 percent and the \(^{207}\text{Pb}/^{206}\text{Pb}\) age of the oldest component may be as great as \((4950 \pm 100)\) million years.

**Summary**

A brief outline of the sequence of events which led to the discoveries of the Pre-Fermi natural reactor and the occurrence of \(^{244}\text{Pu}\) in the early solar system is presented. Great difficulties were encountered in explaining the origin of the so-called CCF (carbonaceous chondrite fission) xenon since the 1960’s. Results from recent studies indicate, however, that this strange xenon component is a mixture of \(^{244}\text{Pu}\) fission xenon and mass-fractionated trapped xenon. A new dating method for the early solar system based on the decay of \(^{244}\text{Pu}\) to the stable xenon isotopes \(^{131}\text{Xe}\), \(^{132}\text{Xe}\), \(^{134}\text{Xe}\) and \(^{136}\text{Xe}\) has recently been established. The \(^{244}\text{Pu}\) ages of meteorites and lunar samples cover a period of nearly one thousand million years, from about 4000 million to almost 5000 million years ago.

**Acknowledgments**—I wish to express my deep gratitude to Professor Koh Sakamoto of Kanazawa University for his very kind introduction; to Professor Kunihiko Watanuki of the University of Tokyo, President of the Geochemical Society of Japan, for bestowing me this highest honor.

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