

Some Neutron and Gamma Radiation Characteristics of Plutonium Cermet Fuel for Isotopic Power Sources

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Gamma and neutron measurements on various types of plutonium sources are presented in order to show the effects of ^{17}O , ^{18}O , ^{19}F , ^{238}Pu , age of the fuel, and size of the source on the gamma and neutron spectra. Analysis of the radiation measurements shows that fluorine is the main contributor to the neutron yields from present plutonium-molybdenum cermet fuel while both fluorine and ^{238}Pu daughters contribute significantly to the gamma-ray intensities.

Plutonium-238 has many desirable qualities for a radioisotopic heat source fuel and has been used in numerous heat sources for terrestrial, marine and space applications. The plutonium fuel has been prepared in several different forms depending on the requirements of the particular mission and the technology available at the time of fabrication. Fuel forms used in the past have been plutonium metal, alloys, and dioxide microspheres. The most recent fuel form to be used is plutonium-molybdenum cermet, (PMC), which is plutonium dioxide that has been coated with molybdenum and hot pressed into the desired shape. This paper deals with the gamma and neutron radiation emitted by the PMC fuel.

The gamma radiation can be divided into three groups; that due to radioactive decay of ^{238}Pu and other nuclides, alpha particle reactions, and fission of the plutonium. Figure 1 shows the gamma spectrum obtained with a Ge(Li) spectrometer of a 15-watt Apollo heat source fueled with plutonium dioxide microspheres. In this spectrum we can see the ^{238}Pu full energy peaks at 100, 153, and 208 keV, several peaks centered around the intense 766 keV peak, and peaks at 851, 883, 926, 942, and 1001 keV. There are some other less intense ^{238}Pu peaks which do not show up in this spectrum.

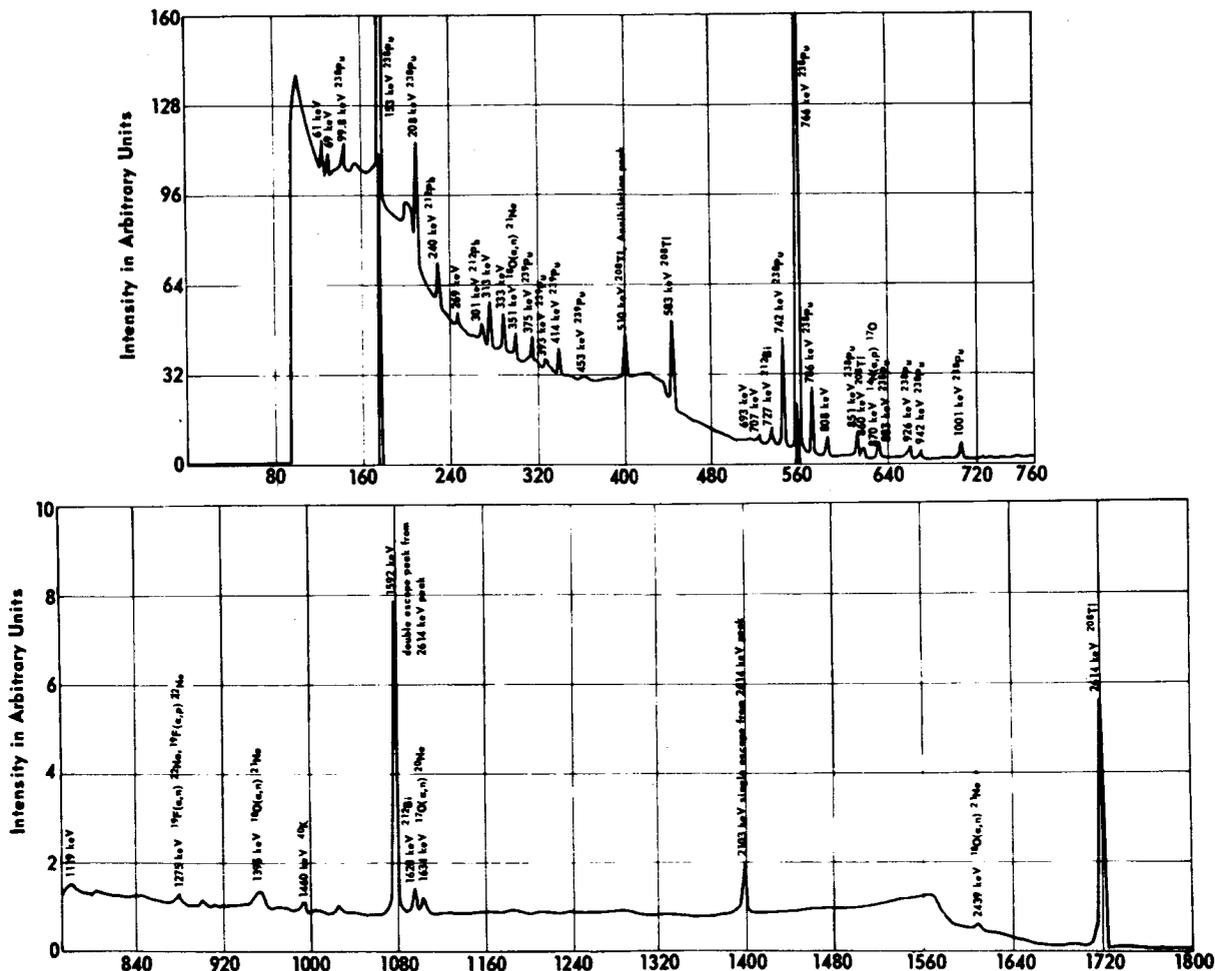
The fuel used in heat sources is nominally about 80% ^{238}Pu , 16% ^{239}Pu , and 1.2 ppm ^{235}Pu . The ^{239}Pu contributes a group of peaks around 400 keV while the main contributions from ^{238}Pu are the two intense peaks at 2614 and 583 keV due to the ^{208}Tl daughter. Other peaks from ^{238}Pu daughters which can be seen in this spectrum are the 510 and 860 keV peaks also from ^{208}Tl , the 240 and 301 keV peaks from ^{212}Pb , and the 727 and 1620 keV peaks from ^{212}Bi . However, a portion of the 510 keV peak is due to annihilation radiation following pair production from the more energetic gamma rays. About five years after chemical separation, the gamma dose rate from the ingrowth of these ^{238}Pu daughters is about equal to the dose rate from the ^{238}Pu gammas for a small encapsulated heat source. The dose rate from these daughters reaches a maximum at 18 years after separation at which time it is ~2.5 times the dose rate from the ^{238}Pu gammas.

Another source of gamma rays is fission, both spontaneous and neutron induced, which will give approximately an exponential continuum of gamma rays. For the Pioneer Program, there was some concern over the gamma rays in the 1 to 4 MeV range. We estimate that for a source the size of the Pioneer there are about 1.8×10^4 photons/sec/watt in the 1-2 MeV

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Figure 1

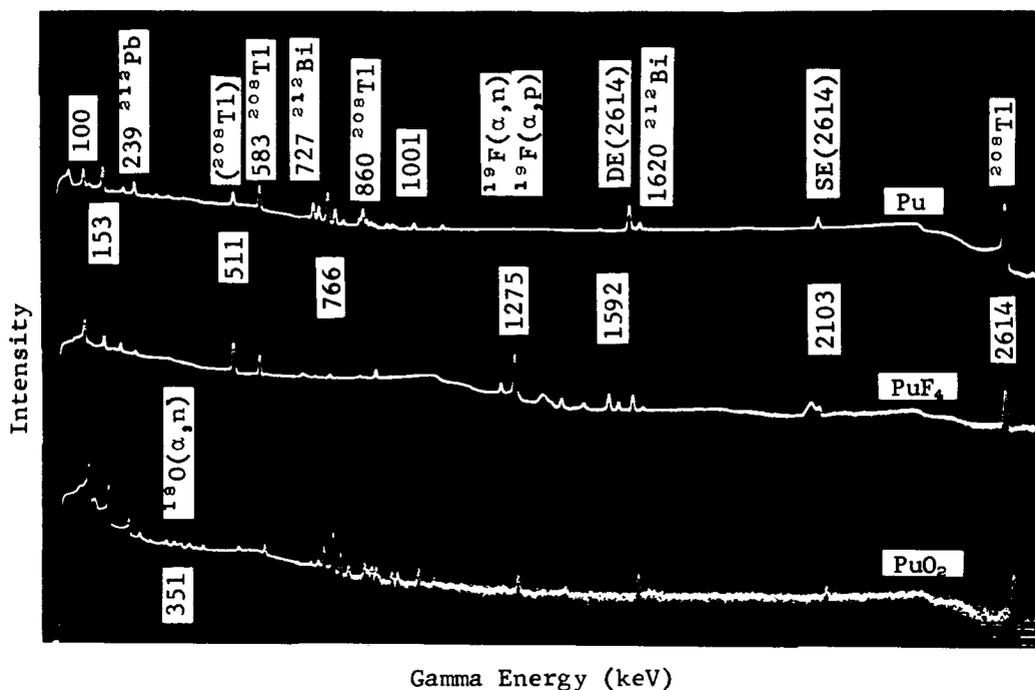
HEAT SOURCE OXIDE FUEL FORM



range due to fission, 6.0×10^3 in the 2-3 MeV range, and 1.9×10^3 in the 3-4 MeV range. These estimates are for bare fuel and have not been corrected for self-absorption within the source. To compare these intensities to the ^{238}Pu photopeaks, the 742, 766, and 786 keV peaks give a total of $\sim 5.1 \times 10^5$ photons/sec/watt from a bare source with no self-absorption, and the total of other ^{238}Pu gamma rays between 880 and 1085 is $\sim 5.4 \times 10^4$ photons/sec/watt.

Figure 2 shows the gamma spectra of three different plutonium sources obtained with a Ge(Li) spectrometer. The top spectrum in this figure is of a metal source containing about seven grams of ^{238}Pu . This spectrum shows the prominent ^{238}Pu peaks (some of which are labeled by energy) as well as the peaks due to ^{238}Pu daughters. Since this source is about seven years old, these peaks from ^{238}Pu daughters are relatively intense.

Figure 2

GAMMA SPECTRA OF Pu, PuF₄, and PuO₂

The bottom curve in Figure 2 shows the gamma spectrum of a PuO₂ source containing about 1.5 grams of ²³⁸Pu. Comparing the top and bottom curves, we can see that the presence of oxygen gives only a few relatively weak gamma rays. These are at 351, 1395, and 2439 keV from ¹⁸O(α,nγ) reactions and at 1634 keV from the ¹⁷O(α,nγ) reaction. For this oxide source, the gamma rays from the ²³⁸Pu daughters are not as intense relative to the ²³⁸Pu gamma rays since the material is much younger than the metal source. However, the ²⁰⁸Tl peaks at 583 and 2614 keV are still prominent.

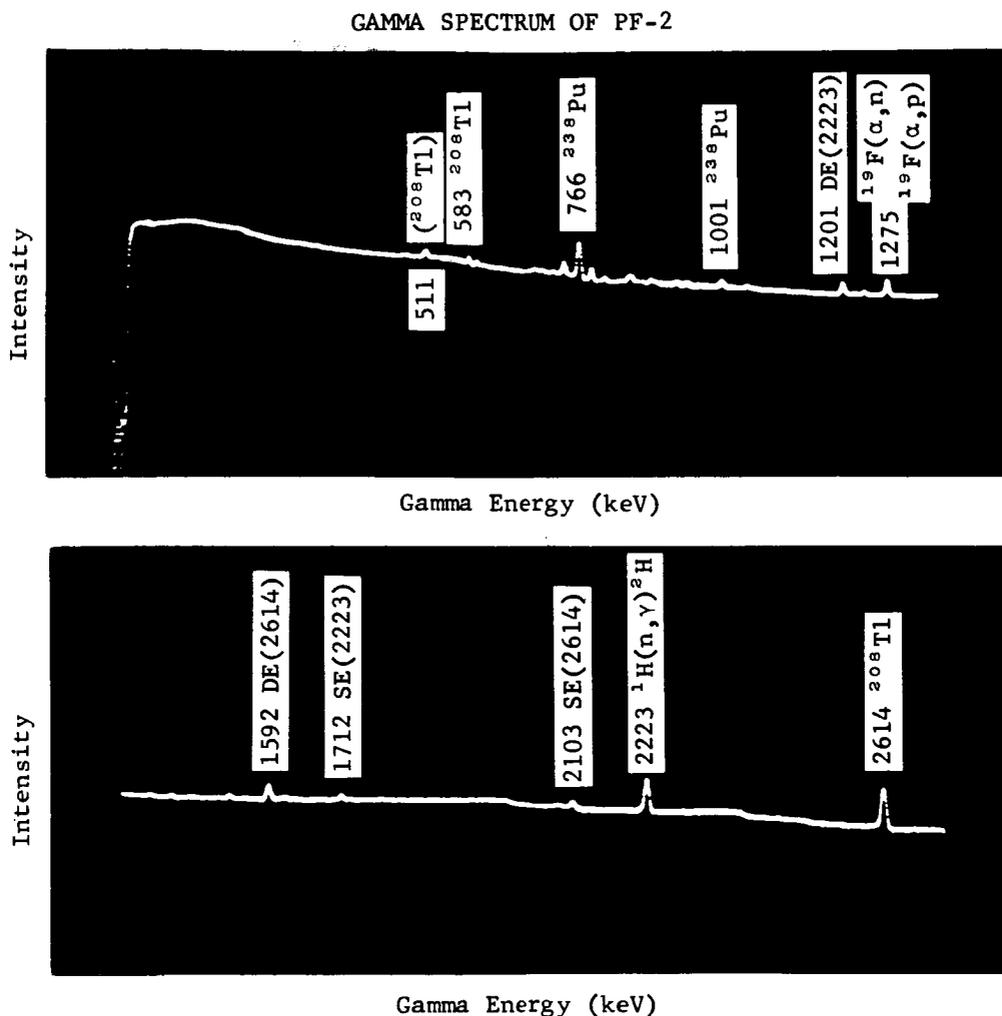
The gamma spectrum of a PuF₄ source containing about 0.5 grams of ²³⁸Pu is shown as the middle curve in Figure 2. The contribution to the gamma spectrum due to fluorine can be seen as a very intense peak at 1275 keV due to both (α,p) reactions on ¹⁹F and the decay of ²²Na formed by the ¹⁹F(α,n)²²Na reaction. There are other peaks due to fluorine at 580, 890, and 2080 keV and five additional peaks clustered around the 1275 keV peak. In normal heat source material, the fluorine content is low enough that the only significant contribution from the fluorine reactions is the peak at 1275 keV.

Since a portion of the 1275 keV peak is due to the decay of ²²Na formed by ¹⁹F(α,n)²²Na reactions, the intensity of this peak will increase with time up to about six years (two half-lives of ²²Na) after the introduction of fluorine. After six years, secular equilibrium is reached and the intensity of the 1275 keV peak remains constant relative to the ²³⁸Pu gamma rays. Measurements have shown that the intensity of the 1275 keV peak will approximately double over this six year period.

Since the PuF₄ source used for Figure 2 was over six years old, the spectrum readily shows the gamma rays from the ²³⁸Pu daughters as well as some of the more prominent ²³⁸Pu peaks.

Shown in Figure 3 is the gamma-ray spectrum of a 647-watt Pioneer capsule fueled with plutonium-molybdenum cermet. Notice the absence of structure below about 500 keV as compared to the spectra shown in Figures 1 and 2. This is due mostly to the greatly increased self-absorption for this source. We still see the ²³⁸Pu group around 766 keV and the 1001 keV peak, also the ²⁰⁸Tl peaks at 583 and 2614 keV, and the fluorine peak at 1275 keV. The peak at 2223 keV is

Figure 3



not from the source but was caused by the $^1\text{H}(n,\gamma)^2\text{H}$ reaction with the water bath the source was in during the measurements.

The neutrons from PMC fuel are contributed by spontaneous fission of ^{238}Pu , neutron induced fission of the Pu, and (α,n) reactions primarily with ^{17}O , ^{18}O , and ^{19}F . Figure 4 shows the neutron spectrum of a 15-watt plutonium metal source. About 75% of the neutrons from this source are from spontaneous fission of the ^{238}Pu while the remaining 25% are mostly due to (α,n) reactions with low Z impurities. Since the neutrons above ~ 5 MeV are almost all due to spontaneous fission, this portion of the spectrum was fitted to a Maxwellian distribution in order to approximate a fission spectrum. The spontaneous fission neutron yield is about 2.65×10^3 n/sec/gram of ^{238}Pu .

This value is represented by the lower curve in this figure and is the minimum number of neutrons that a ^{238}Pu source can emit. The average neutron energy for this fission curve is 2.0 MeV.

Even though PMC fuel is enriched in ^{18}O which will not undergo (α,n) reactions with the 5.5 MeV ^{238}Pu alpha particles, there are still sufficient quantities of ^{17}O and ^{18}O present to produce significant quantities of neutrons. The effects of ^{17}O and ^{18}O on the neutron spectrum are shown in Figure 5. This is the spectrum of a PuO_2 source in which the oxygen is enriched to 45.6% ^{18}O and 1.7% ^{17}O whereas natural oxygen is 0.2% ^{18}O and 0.04% ^{17}O . The data at energies greater than 1 MeV were obtained with a stilbene spectrometer while the datum point between 0 and 1 MeV was obtained from a difference calculation

the peak at about 1.25 MeV. The average neutron energy for this source is 1.2 MeV, and the specific yield is 2.8×10^6 n/sec/gram of ^{238}Pu .

Figure 4
NEUTRON ENERGY SPECTRUM OF SOURCE HP-15-2

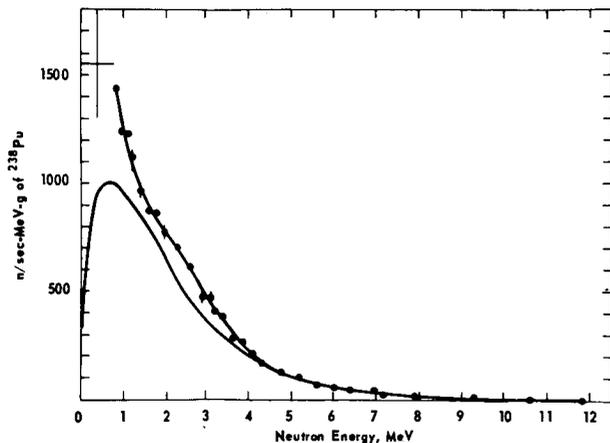
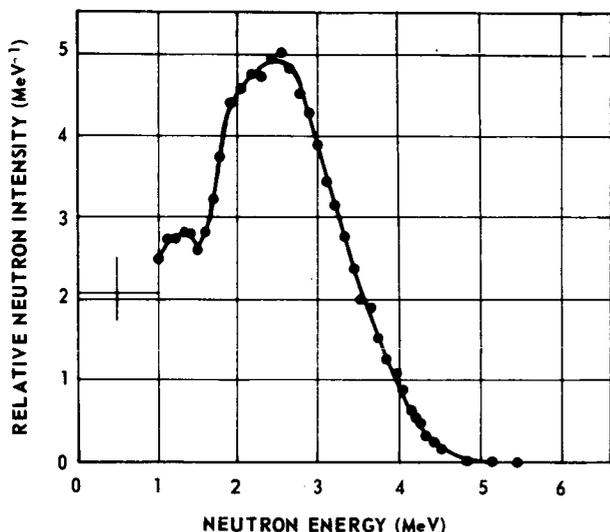


Figure 5

NEUTRON ENERGY SPECTRUM OF A ^{238}Pu - ^{18}O (α ,n) SOURCE

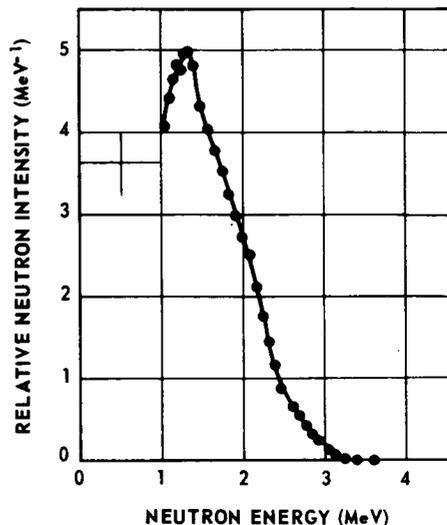


using the stilbene data and a long counter measurement. In this spectrum, we see a broad maximum around 2.5 MeV due to the (α ,n) reactions with ^{17}O and ^{18}O . These (α ,n) reactions yield about 1.4×10^4 n/sec/gram of ^{238}Pu in a PuO_2 source with oxygen of natural isotopic abundance and give a maximum neutron energy of about 4.5 MeV. The average neutron energy from this source is 2.1 MeV.

A similar spectrum only of a PuF_4 source is shown in Figure 6. The maximum energy the neutrons from the $^{19}\text{F}(\alpha$,n) ^{22}Na can have, using a 5.5 MeV alpha particle, is 3.2 MeV. This (α ,n) reaction produces

Figure 6

NEUTRON ENERGY SPECTRUM
OF A ^{238}Pu - ^{19}F (α ,n) SOURCE



In Figure 7 we can see the effect of the neutrons due to neutron induced fission of the plutonium. The lower curve is a log plot of the data for the 15-watt metal source that was shown in Figure 4 as a linear plot. The upper curve is the neutron spectrum of a 1482 watt SNAP-27 source. Because the SNAP-27 was a natural oxygen PuO_2 source, there is a broad maximum around 2.5 MeV from the $\text{O}(\alpha$,n) reactions. Since both of these spectra are plotted as specific yield per MeV, the spontaneous fission spectrum for the metal source also represents the spontaneous fission curve for the SNAP-27 source. At neutron energies greater than 5 MeV, the neutron induced fission spectrum is represented by the difference between the two curves. For this SNAP-27 source, the neutron yield from induced fission was $\sim 3.7 \times 10^3$ n/sec/gram of ^{238}Pu . This gave a neutron multiplication in this source of about 20%. The total specific yield for this source was 2.2×10^4 n/sec/gram of ^{238}Pu , and the average neutron energy was 1.9 MeV.

Figure 7

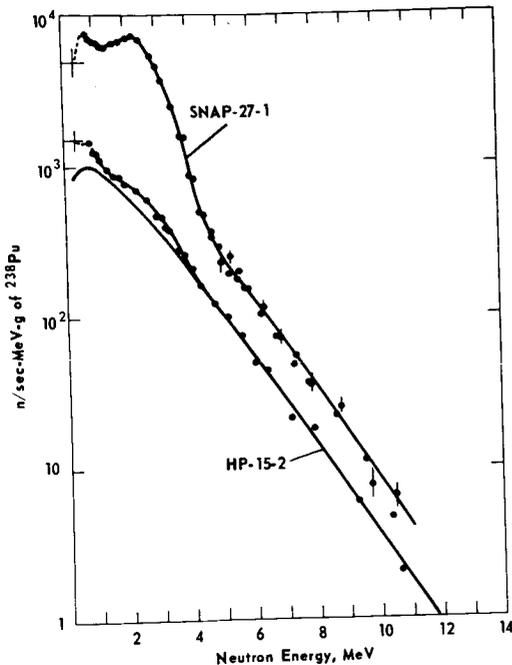
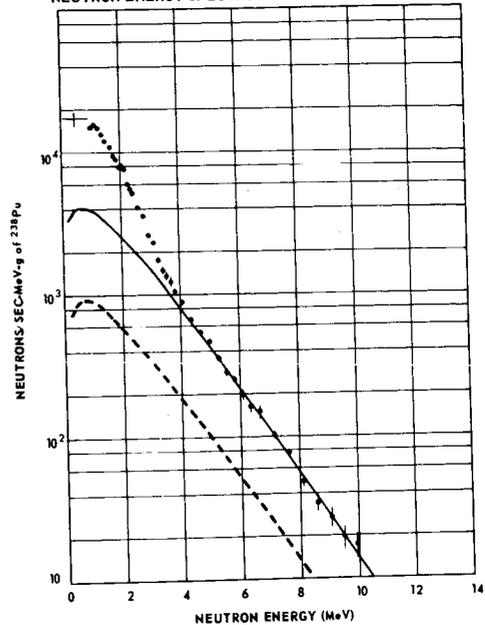
ENERGY SPECTRA OF TWO ^{238}Pu POWER SOURCES

Figure 8

NEUTRON ENERGY SPECTRUM OF PIONEER CAPSULE PF-2



Shown in Figure 8 is the neutron spectrum of the 647-watt, PMC fueled, Pioneer capsule PF-2. The dashed line represents the spontaneous fission curve, the solid line is the sum of the spontaneous fission and induced fission curves, and the data points represent the measured neutron spectrum. The rise in the spectrum below 5 MeV is due to the presence of ^{17}O and ^{18}O which remained after the ^{16}O exchange process and to fluorine that was introduced into the fuel during the molybdenum coating process. This source had a specific yield of 3.73×10^4 n/sec/gram of ^{238}Pu . It is estimated from the spectral measurements that approximately 7% of these neutrons were due to spontaneous fission of the ^{238}Pu , 7% to $\text{O}(\alpha, n)$ reactions, 23% to induced fission, and 63% to the $^{19}\text{F}(\alpha, n)^{22}\text{Na}$ reaction. Neutron measurements on individual PMC discs that were stacked together to make a Pioneer capsule and on the finished capsule show an increase due to multiplication of 26%. The average neutron energy for this source was 1.3 MeV.

Table I contains a summary of the neutron data. From this table, we can see how prolific the oxygen and especially the fluorine are for producing neutrons. We can also see the effects of these two elements on the average energy and the dose conversion factors.

In summary, it is evident that the main problem, as far as lowering neutron levels in PMC fuel, is the fluorine content. Most likely, future PMC fuel will be produced using MoCl_5 for the molybdenum coating rather than MoF_6 . This would keep all but trace amounts of fluorine out of the fuel. The small neutron contribution from ^{17}O and ^{18}O is being still further reduced through improved ^{16}O exchange methods.

For the gamma radiation, the fluorine is again a problem along with the ingrowth of ^{236}Pu daughters. It may be possible in the future to obtain fuel with a lower ^{238}Pu content or to age the fuel to allow some of the ^{238}Pu to decay before performing chemical separation.

Table 1

SUMMARY OF NEUTRON DATA

SOURCE	SPECIFIC YIELD n/sec/g ^{238}Pu	AVERAGE NEUTRON ENERGY	CONVERSION FACTORS FROM NEUTRON FLUENCE TO	
			DOSE EQUIVALENT	ABSORBED DOSE
Fission*	2.6×10^3	2.0 MeV	3.3×10^{-8} Rem/n/cm ²	3.8×10^{-9} Rad/n/cm ²
$^{238}\text{PuO}_2$	1.8×10^4	2.1	3.4	4.0
$^{238}\text{PuF}_4$	2.8×10^4	1.2	3.1	3.3
SNAP-27	2.2×10^4	1.9	3.3	3.8
PF-2	3.7×10^4	1.3	3.1	3.4

*Calculated