LETTERS TO THE EDITOR

$^{238}PuO_2$ AS A HEAT SOURCE FUEL

Because of the present and potential use of $^{238}PuO_2$ as a heat source fuel, I wish to point out that the neutron radiation levels for fuel currently being produced are considerably lower than those given in the recent article by Taherzadeh and Gingo, "Neutron Radiation Characteristics of Plutonium Dioxide Fuel," *Nucl. Technol.*, **15**, 396 (1972). One reason for this is that when it is important to minimize neutron levels, PuO_2 which has been depleted in ^{17}O and ^{18}O is used for the fuel, whereas in this article the PuO_2 is assumed to contain oxygen of natural isotopic abundance. Oxygen-17 and ^{18}O are the naturally occurring isotopes of oxygen which can undergo (α, n) reactions with alpha particles from ^{238}Pu. The specific neutron yield from the $O(\alpha, n)$ reaction, only, for fuel produced at Mound Laboratory which has been depleted in these two isotopes has been between 3×10^2 and 3×10^3 n/(sec g) ^{238}Pu, depending on the degree of depletion. This compares with a measured yield of 1.4 to 1.5×10^4 n/(sec g) ^{238}Pu from the $O(\alpha, n)$ reaction for undepleted PuO_2. The authors have calculated a yield of $1.99 \pm 0.45 \times 10^4$ n/(sec g) ^{238}Pu [or $(1.42 \pm 0.32) \times 10^4$ n/(sec g) (PuO_2-80% ^{238}Pu)] from the $O(\alpha, n)$ reaction for undepleted PuO_2.

Another reason for the neutron levels in the article being higher than current practice is the relatively high concentrations of low Z impurities assumed by the authors. Based on these concentrations, they have calculated a yield of 1.3×10^4 n/(sec g) ^{238}Pu for (α, n) reactions with impurities. For fuel produced recently at our laboratory, the neutron levels from (α, n) reactions with impurities have been $\leq 2 \times 10^3$ n/(sec g) ^{238}Pu.

The authors have calculated the total specific yield for a particular 2200-W(th) source fueled with undepleted PuO_2. Their value, which includes neutron-induced multiplication, is 6.42×10^4 n/(sec g) ^{238}Pu [or 4.59×10^4 n/(sec g) PuO_2]. Using the same source design but assuming the PuO_2 is depleted in ^{17}O and ^{18}O, as is currently being produced for large size heat sources, the expected total specific yield, including multiplication, would be only about 7 to 8×10^3 n/(sec g) ^{238}Pu.

Unfortunately, the article contains some errors. Two which might result in confusion for some readers are

1. *Section IIIA.* "The gamma-ray energy emitted from the excited states of the daughter nucleus ($^{234*}U$) has a maximum energy of 43.5 keV, and thus will be absorbed by the source or the shield surrounding it." Gamma rays as high as 1085 keV from the daughter nucleus have been observed, and these will not all be absorbed.
2. *Section IV.* There is the suggestion that 5.11×10^{11} might be the alpha yield per second per gram of ^{238}Pu. In the reference this value came from, 5.11×10^{11} is stated as being the yield per gram of ^{238}Pu product, which most likely means per gram of total plutonium of which 81% is ^{238}Pu.

M. Edward Anderson

Monsanto Research Corporation
Mound Laboratory
Miamisburg, Ohio 45342

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REPLY TO $^{238}PuO_2$ AS A HEAT SOURCE FUEL

The author of the letter has stated that the experimental value of the neutron yield of the *current* Mound Laboratory plutonium dioxide fuel heat source is considerably lower than the reported value[1] in *Nuclear Technology*.

1. The neutron emission characteristics of plutonium dioxide fuel were estimated in Ref. 1 for a typical fuel source available at the time the report was being prepared. The difference between the heat sources selected by Ref. 1 and by Mound Laboratory is in the quantity of the low-Z impurities and ^{16}O enrichment. Obviously, if a low impurity and ^{17}O-^{18}O depleted source is used, a lower neutron yield would be expected.

In general, one can estimate the neutron emission yield of any plutonium dioxide fuel, including the current Mound Laboratory fuel, by using the *Nuclear Technology* report, provided the characteristics of the fuel are made available. Disregarding the difference between the two heat sources under question, there is good agreement between the theoretical calculation $(1.99 \pm 0.45) \times 10^4$ n/(sec g) ^{238}Pu (Ref. 1) and experimental estimated value of $(1.54 \pm 0.07) \times 10^4$ n/(sec g) ^{238}Pu (Mound Laboratory, Letter to the Editor). These numbers are in good agreement. An analytical calculation is based on the integration of the $O(\alpha, n)$ reaction rate, and the parameters involved had to be gathered from various publications. The magnitude of the reaction cross section, the

decay probabilities of the ^{21}Ne excited states, the $^{18}O/^{16}O$ ratio, the magnitude of plutonium alpha yield, and the energy loss equation of the heavy charged particles in the very low energy range were all obtained from the literature. In addition, assumptions were made to simplify the calculations, such as the isotropic angular distribution of the neutrons in the center-of-mass system. The two-body kinematics and the compound nuclear model were also chosen for the neutron yield estimation. Considering these assumptions, the agreement between the two values given above is quite satisfactory.

2. The fact that the *current* Mound Laboratory fuel is depleted in ^{17}O and ^{18}O, and has less low-Z impurity does not change the conclusion or the results of the report published in *Nuclear Technology*. Mound Laboratory should be commended for their preparation of a higher quality fuel and for providing the latest value of 7 to 8×10^3 n/(sec g) ^{238}Pu measured experimentally for their current fuel. However, to indicate the huge difference between the two results without referring to the obvious reasons is misleading. In contrast, our analytical value of 6.42×10^4 and the Mound experimental value of 7000 to 8000 do indeed agree within the predicted uncertainties if the differences between the two fuels are incorporated in the results.

One can start with the analytical value of 6.42×10^4 and correct for the characteristics of the Mound Laboratory heat source:

TABLE I

Fuel Characteristics Used by Ref. 1 and Mound Laboratory

Characteristics	Ref. 1	Mound Laboratory
Fuel enrichment	0.204%	Much less[a] (exact value not given)
Multiplication factor	1.55	1.30
Impurities	Table II, p. 407	Much less[b] (exact value not given)
Neutron yield due to impurities	$(1.29 \pm 0.02) \times 10^4$ n/(sec g) ^{238}Pu	2000 ± 700 n/(sec g) ^{238}Pu

[a]The value of the fuel enrichment was not given by the author. From other documents we can estimate approximately one-third.

[b]The quantities of the low-Z impurities of Mound Laboratory plutonium fuel are not known.

a. Use the correct ^{16}O enrichment fuel (Table I). The *Nuclear Technology* report used natural oxygen; therefore,

$$(6.42 \times 10^4)\left(\tfrac{1}{3}\right) = 2.14 \times 10^4 \quad .$$

b. Use the correct value of the subcritical multiplication factor (Table I); therefore,

$$(2.14 \times 10^4) \times (1.3/1.55) = 1.795 \times 10^4 \quad .$$

c. Correct for the difference in the low-Z impurities contamination (Table I); therefore,

$$(1.795 \times 10^4) - (1.3 \times 10^4) + (2 \times 10^3) = 6.95 \times 10^3 \quad .$$

The correct comparison is made when the number 6.95×10^3 is compared with 7000 to 8000. Clearly, a more accurate analytical value can be estimated if the ^{16}O enrichment and the quantity of the low-Z impurities are made available.

3. The 43.5 keV was the maximum energy of the gamma rays with the abundances >0.03%, and they are generally absorbed by the medium. Gamma rays of higher energies are emitted from $^{234*}U$ but with extremely low abundance. In any case, these gamma rays are not contributing to the neutron yield of the source, and thus it is an irrelevant fact to the report.

4. The value of 6.35×10^{11} alpha/(sec/g) ^{238}Pu was used throughout the report and for the final analysis. The author is correct in pointing out that 5.11×10^{11} is alpha/(sec g) Pu.

Conclusion: I believe the agreement between the experimental value of the neutron yield from a plutonium dioxide fuel and the analytical value is a good indication that the analytical approach to the problem was the correct one. In addition, analytical results could have been used to estimate the neutron yield of the current Mound Laboratory plutonium dioxide fuel heat source.

Mojtabe Taherzadeh

Jet Propulsion Lab
California Institute of Technology
4800 Oak Grove Drive
Pasadena, California 91103

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REFERENCES

1. M. TAHERZADEH and P. GINGO, "Neutron Radiation Characteristics of Plutonium Dioxide Fuel," *Nucl. Technol.*, **15**, 396 (1972).