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LLNL-TR-438648

U232 Nonproliferation Features

R. Moir

June 25, 2010

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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Ralph Moir

June 23, 2010

Vallecitos Molten Salt Research
607 E. Vallecitos Rd.
Livermore, CA 94550
925-447-8804 phone/fax
925-413-6131 cell
ralph@ralphmoir.com
www.ralphmoir.com/

Abstract

U232 (^{232}U) as a contaminant in U233 (^{233}U) can play a role in nonproliferation of nuclear weapons and still allow U233 to be used in the nuclear fuel cycle. This note describes the two features of U232, its gamma radiation and its heat release, both of which causes problems for use in nuclear weapons. The gamma radiation at low levels causes health problems and even death for people nearby after prolonged exposures. At high levels it causes degradation of the high explosives of the weapons after a sufficiently long exposure. The heat causes problems in weapons design, for example, thermal degradation of high explosives. We quantify these properties of U232 in this note. The role of U232 as a potential nuclear weapon contaminant is similar to that of Pu238 (^{238}Pu) in that both give off troublesome heat, U232 much more than Pu238, but the strong radiation from U232 makes its role unique as Pu238 has little radiation associated with it.

The gamma dose to a worker at 1 m from 5 kg of U233 with 2.4% U232 would be 100 rem/hr, 1 year after chemical separation. The dose would be 4 rem after 65 hours of exposure, the limit for annual exposure to workers and a fatal dose of 300 rem would occur in 300 hours of exposure after separation. The high explosive is predicted to degrade owing to ionizing radiation after a little over ½ year. The heat rate is 77 W just after separation and climbs to over 600 W ten years later.

In this note we first discuss the effects of gamma radiation from U232 on personnel and the effect on damage to high explosives. Then we discuss the heat generation rate from decay of U232. Finally we discuss the temperature rise caused by the heat generation rate.

The decay chain of Pu238 whose half-life is 87.7 years has eight alpha decays and four beta decays to Pb206. However, its first daughter is U234 with a 245,500-year half-life causing the decay chain to stop there for practical purposes. The energy release is 5.59 MeV, typically with a 5.5 or 5.46 MeV alpha and 99.9 keV gamma.

By contrast, the decay chain of U232 (Fig. 1) whose half-life is 70 years has six alpha decays and two beta decays to stable Pb208. The energy release in the first step with a half-life of 70 years to Th228 is 5.4 MeV. There is holdup in the next decay of Th228 with its 1.9-year half-life. The remaining decays take place with short half-lives with a total decay energy of 40.8 MeV including a penetrating gamma ray of 2.6 MeV and some gamma radiation of much lower energy. That is, the heat generation rate increases up to a factor of (40.8/5.4=) 7.6 with time. There would be a small correction for the energy carried away by neutrinos that we neglect here.

The heat coming from U232 would be 1.2 times that of Pu238 for the same mass, where NU232 is the number of U232 atoms in a given mass and τ_{232} is the decay time constant.

$$\left(\frac{NU232}{NPu238}\frac{\tau_{238}}{\tau_{232}}\frac{E_{232}}{E_{238}}\right)=\left(\frac{238}{232}\frac{87.7}{70}\frac{5.4}{5.59}\right)=1.24$$

until the Th228 builds up after which time the heating would be 9.4 times more

$$\left(\frac{NU232}{NPu238}\frac{\tau_{238}}{\tau_{232}}\frac{E_{232\,total}}{E_{238}}\right)=\left(\frac{238}{232}\frac{87.7}{70}\frac{40.8}{5.59}\right)=9.38$$

and have 33 times more gamma radiation energy that is much more penetrating.

$$\left(\frac{NU232}{NPu238}\frac{\tau_{238}}{\tau_{232}}\frac{E_{\gamma 232}}{E_{\gamma 238}}\right)=\left(\frac{238}{232}\frac{87.7}{70}\frac{2.6}{0.1}\right)=33.4$$

HEAT AND RADIATION GENERATION RATE

The rate of release of energy is

$$heat\,rate=\frac{NU232}{\tau_{232}}E_{232}+\frac{NTh228}{\tau_{228}}(E_{total}-E_{232)}\text{ in units of MeV/y per atom of U232.}$$

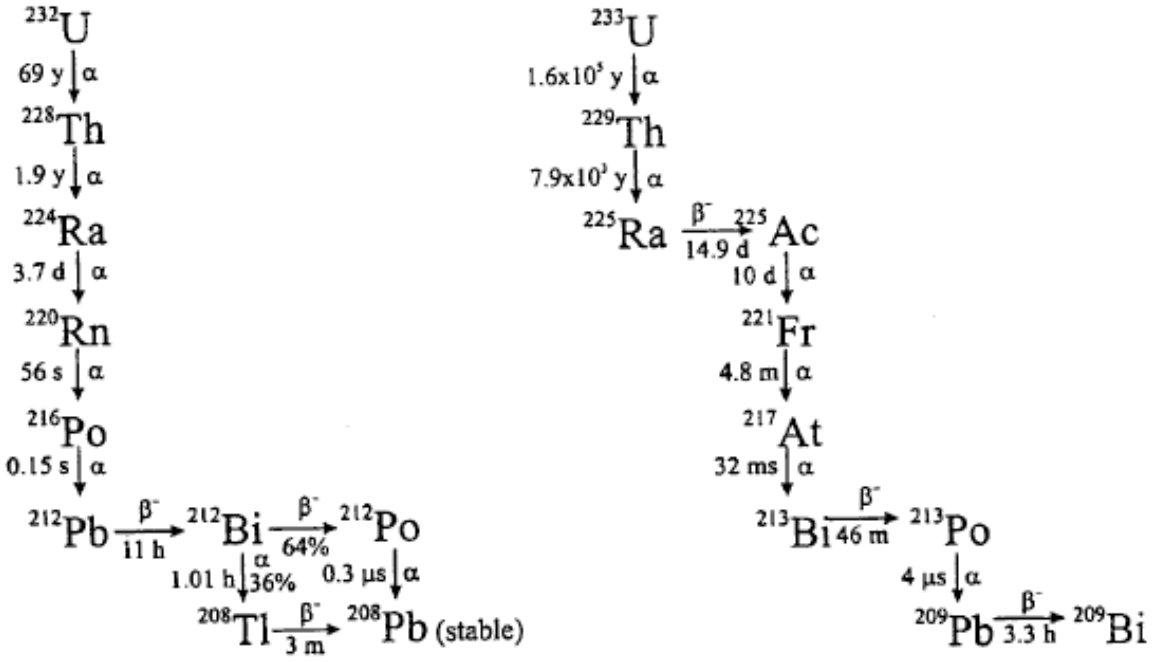

Fig. 1. The decay chains of U232 and U233.

If we multiply by

$$\left(\frac{M(kgU232)}{232.03713\times 1.66054\times 10^{-27}\,kg/atom}\right)\times\left(\frac{1.6021\times 10^{-19}\,j/eV\times 10^{6}eV/MeV}{365.25\times 24\times 3600 s/y}\right)=13{,}180\,W/kg$$

we get units of W.

$$P=\left(heat\ rate\ in\ MeV/atom U232\bullet year\right)\times\left(M(kgU233)\right)\left(\frac{NU232}{NU233}\right)\times\left(13{,}180\,W/kg\right)+0.28\,W/kg\times\frac{U233}{U}$$

$$E_{232}=5.4\,MeV \qquad E_{total}=40.8\,MeV$$

$$NU232=NU232_0 e^{-t/\tau_{232}} \qquad \tau_{232}=\tau_{1/2}/0.693=70\ years/0.693=101\ years$$

$$\frac{dNTh228}{dt}=\frac{NU232}{\tau_{232}}-\frac{NTh228}{\tau_{228}} \qquad \tau_{228}=\tau_{1/2}/0.693=1.9\ years/0.693=2.74\ years$$

$$NTh228=\int\frac{dNTh228}{dt}dt=\frac{NU232_0}{\left(\frac{\tau_{232}}{\tau_{228}}-1\right)}\left[e^{-t/\tau_{232}}-e^{-t/\tau_{228}}\right]$$

$$heat\ rate=\frac{NU232_0}{\tau_{232}}E_{232}e^{-t/\tau_{232}}+\frac{NTh228}{\tau_{228}}(E_{total}-E232)$$

$$gamma\ rate = \frac{NTh228}{\tau_{228}} E_{gamma} \qquad E_{gamma} \approx 2.6\ MeV$$

If we wish to resolve the gamma production rate in the first week or so after separation of the uranium from other elements while the gamma rate is low we need to include the series step of holdup by Ra224. In this case we need one more differential equation.

$$\frac{dNRa224}{dt} = \frac{NTh228}{\tau_{228}} - \frac{NRa224}{\tau_{224}} \qquad \tau_{224} = \tau_{1/2}/0.693 = 3.7\ d/0.693 = 0.0146\ years$$

$$gamma\ rate = \frac{NRa224}{\tau_{224}} E_{gamma} \qquad E_{gamma} \approx 2.6\ MeV$$

For times >>3.7 d $\qquad \frac{dNRa224}{dt} \approx 0$

$$\frac{NRa224}{NTh228} = \frac{\tau_{224}}{\tau_{228}}$$

The heat rate and gamma rate are shown in Fig. 2. They are based on one atom of U232. We assume at time zero the U232 has just been separated and therefore the Th228 content is zero. Notice that the heat rate is finite at the beginning but the gamma rate starts at zero while the Th228 builds up to a peak in about nine years. The gamma rate peaks at 0.023 MeV/y and the heating peaks at 0.39 MeV/y.

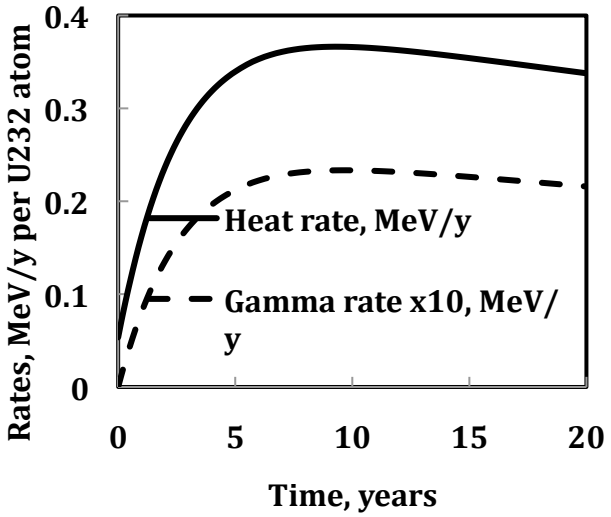

Fig. 2a. Gamma rate in Mev per atom of U232.

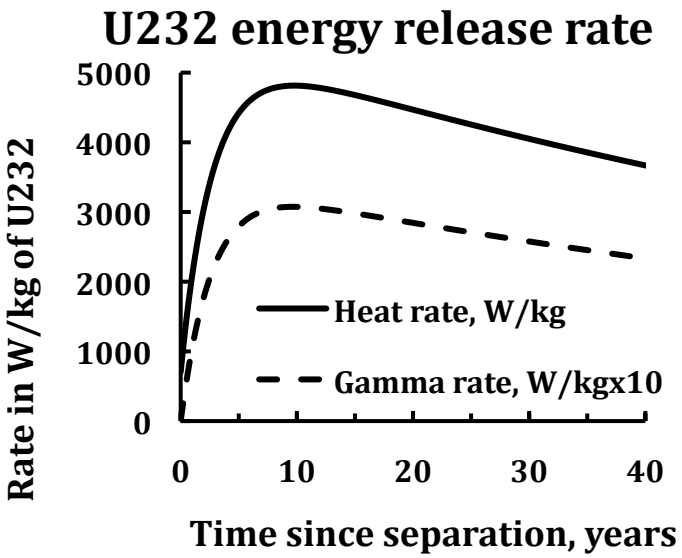

Fig. 2b. Heat rate of U232 in watts per kg.

The half-life of U233 is 159,000 years and its energy release is 4.9 MeV. The heat rate of pure U233 is 0.28 W/kg as is shown:

$$P = \left(heat\ rate\ in\ MeV/atom U233 \bullet year\right) \times \left(\frac{M(kgU233)}{233.04 \times 1.66054 \times 10^{-27} kg/atom}\right)$$

$$\times \left(\frac{1.6021 \times 10^{-19} j/eV \times 10^{6} eV/MeV}{365.25 \times 24 \times 3600 s/y}\right)$$

$$= \frac{4.9\ MeV/atom}{159{,}000\ y/0.693}\left(\frac{1}{233.04 \times 1.66054 \times 10^{-27} kg/atom}\right) \times \left(\frac{1.6021 \times 10^{-19} j/eV \times 10^{6} eV/MeV}{365.25 \times 24 \times 3600 s/y}\right)$$

$$= 0.28\ W/kg$$

The ratio of the heat rate of U233 to pure U232 is

$$\frac{\frac{N233}{\tau_{233}} E_{233}}{\frac{N232}{\tau_{232}} E_{232}} = \frac{N233\ E_{233}\ \tau_{232}}{N232\ E_{232}\ \tau_{233}} = \frac{N233\ \ 4.9\ MeV\ 70\ y}{N232\ \ 5.4\ MeV\ 159{,}000\ y} = 4 \times 10^{-4} \frac{N233}{N232}$$

The heating from pure U233 is 4×10^{-4} times smaller than that of the same amount of

U232 but as Th228 builds up it drops to 5.3×10^{-5} times smaller.
As mentioned before, the amount of U232 to produce the same heat as that of Pu238 is 9.4 times less after 9 years of build up.
The heat rate would double from 0.28 to 0.56 W/kg for $\frac{N233}{N232} = 4 \times 10^{-4} = 400\ ppm$

After nine years the heat rate would be the same 0.56 W/kg at 53 ppm of U232.
As mentioned before the heat rate of U232 is like that of Pu238 on steroids!

$$heat\ rate/kg\ of\ U232 \div heat\ rate\ of\ Pu238 =$$

$$\frac{238}{232}\left[\frac{NU232}{\tau_{232}}E_{232}+\frac{NTh228}{\tau_{228}}(E_{total}-E_{232})\right]\Bigg/\left[\frac{NPu238}{\tau_{238}}E_{238}+\frac{NTh228}{\tau_{228}}E_{238}\right]$$

RADIATION DAMAGE TO HIGH EXPLOSIVE (HE)

The high explosive HMX commonly used in nuclear explosives can withstand up to 1.0×10^{8} r (Avrami, Jackson, and Kirshenbaum, 1973). The effects of this radiation dose are gas evolution, crumbling and other undesirable effects. An r (roentgen) is equal to 0.00877 J/kg. 100 rad=1 gray (Gy) = 1 J/kg. 1 r = 0.877 rad = 0.00877 gray. 100 rem = 1 Sv. For our purposes a rad, a rem and an r are pretty closely equal for gamma radiation.

We now discuss the consequences of various levels of U232/U233 on gamma dose rate from a sphere of U233 of 5 kg reflected by beryllium that would be just critical. At U232/U233 = 0.024 the dose rate at 1 m is 100 rem/h after 1 year from separation (Kang and von Hipple, 2001). We have normalized the dose rate of Fig. 2 to 100 rem/h at 1 year and plotted the result in Fig. 3.

At 0.04 m (contact) the dose rate would be $100/0.04^{2} = 6.3\text{x}10^{4}$ rad/h assuming a rem=rad. High explosive can tolerate about 100 Mr before degradation. $10^{8}/6.3\text{x}10^{4} =$ 1600 hours to accumulate the tolerable dose for 1 year after separation of U232. At nine years the dose rate is 2.9 times that at 1 y. The time to degrade or shelf life would be 550 hours (Fig. 4).

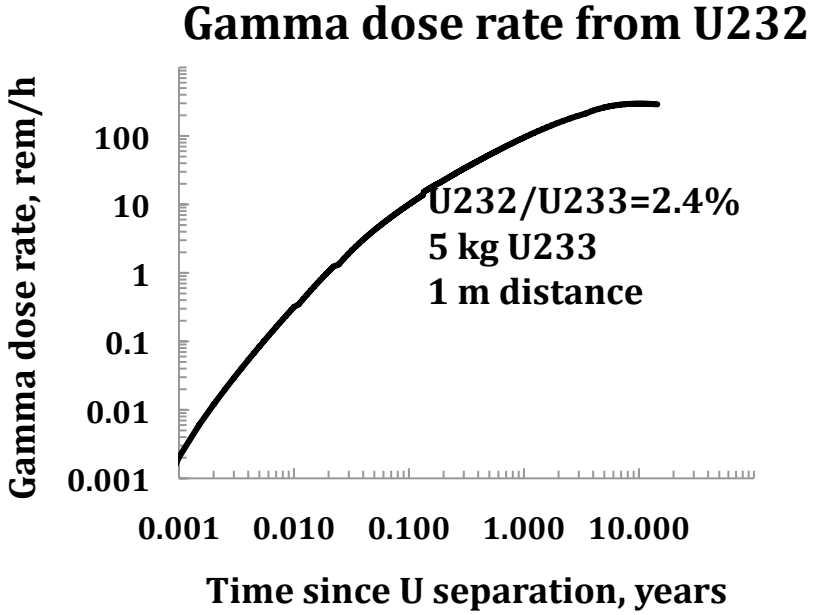

Fig. 3. Gamma dose rate from 5 kg of U233 with 2.4% U232.

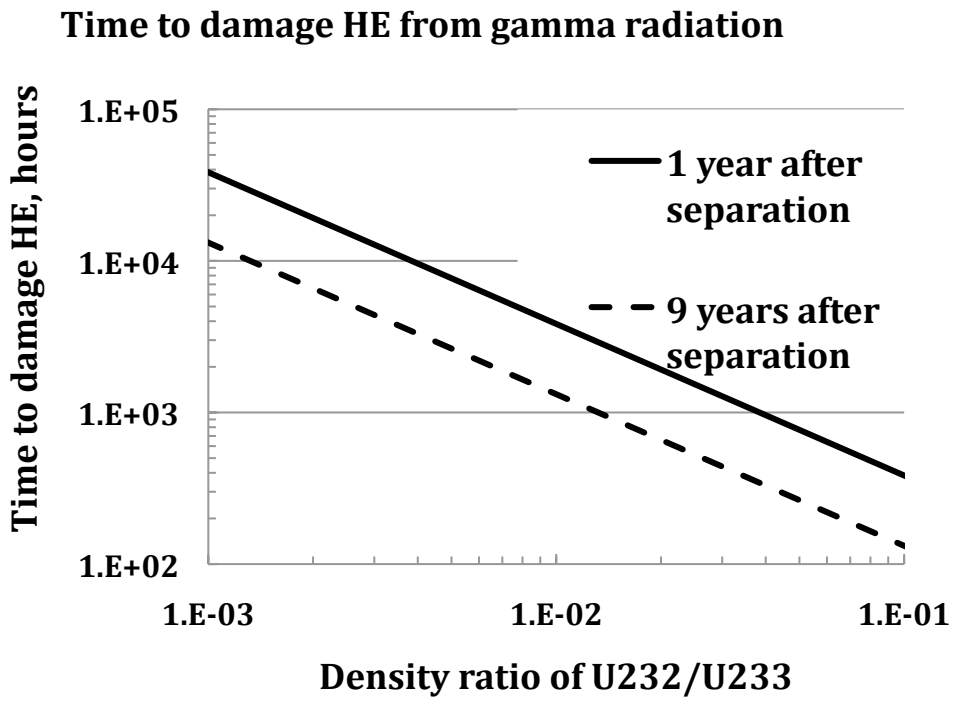

Fig. 4. Time to damage HE from gamma radiation.

The sense of the Fig. 4 is that the life of a nuclear explosive might be about a year at a U232/U233 ratio of 0.01 and to get 10 years life time the ratio would have to be less than about 0.001. To have the HE damage in a month would take a U232/U233 ratio of about 0.05. The heat rate is likely to also be a serious limitation and will be discussed later in the section, *Effects of heat generation*.

A more proper way to assess the dose required to damage HE is to integrate the function of Fig. 3, which is shown in Fig. 5, 6 and 7.

$$\text{Dose}=\int_0^t \frac{NRa224}{\tau_{224}} E_{gamma}\, dt$$

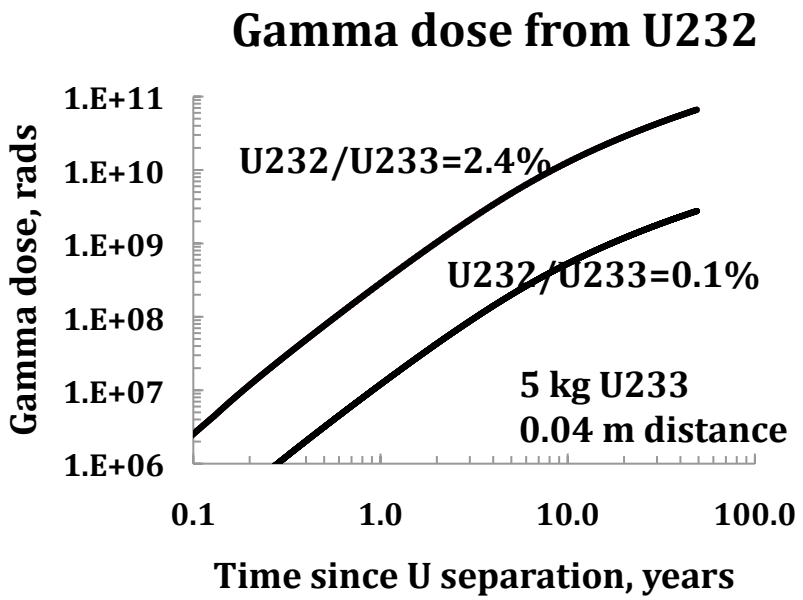

Fig. 5. Contact dose in rads from 5 kg with 0.1% & 2.4% U232 versus time.

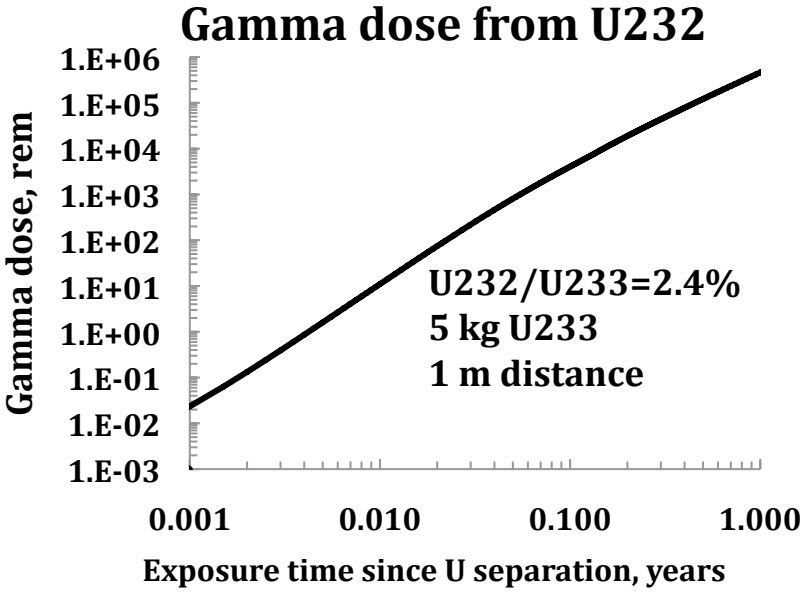

Fig. 6. Dose at 1 m in rems from 5 kg with 2.4% U232 versus time in years.

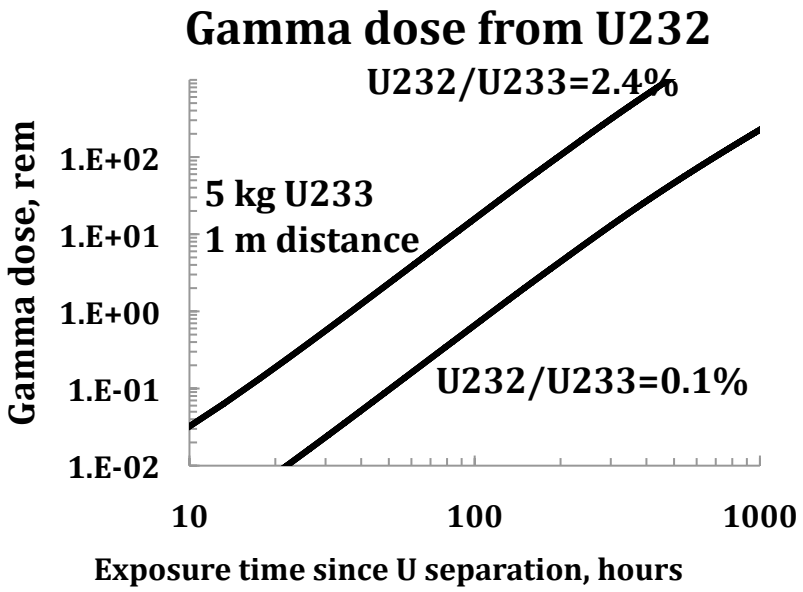

Fig. 7. Dose at 1 m in rems from 5 kg with 0.1 & 2.4% U232 versus time in hours.

The HE damages in 3.4 and 0.58 years for 0.1 and 2.4% U232 from Fig. 5. The U232 concentration ratio is proportional to gamma dose or damage for a fixed time.

$$\frac{NU232}{NU} = 0.024 \frac{damage}{10^8\ rads} \qquad for\ t = 10\ y$$

A ten year shelf life would require the U232/U ratio < 0.00024 or 240 ppm and such a short shelf life might still be of interest to a military. A terrorist might "live" with a short shelf life, perhaps 60 days. Then the U232/U ratio < 0.25, which is an impractically high ratio and so is no limit at all and the heat generated would be limiting at lower concentrations as discussed later in the section, *Effects of heat generation*.

The time to reach a fatal dose of 300 rem is 1135 & 300 hours for 0.1 & 2.4 % U232 from Fig. 7.

From Fig. 7 of Kang and von Hipple (2001) we get the critical mass when reflected with 4-cm thick layer of beryllium shown as Fig. 8:

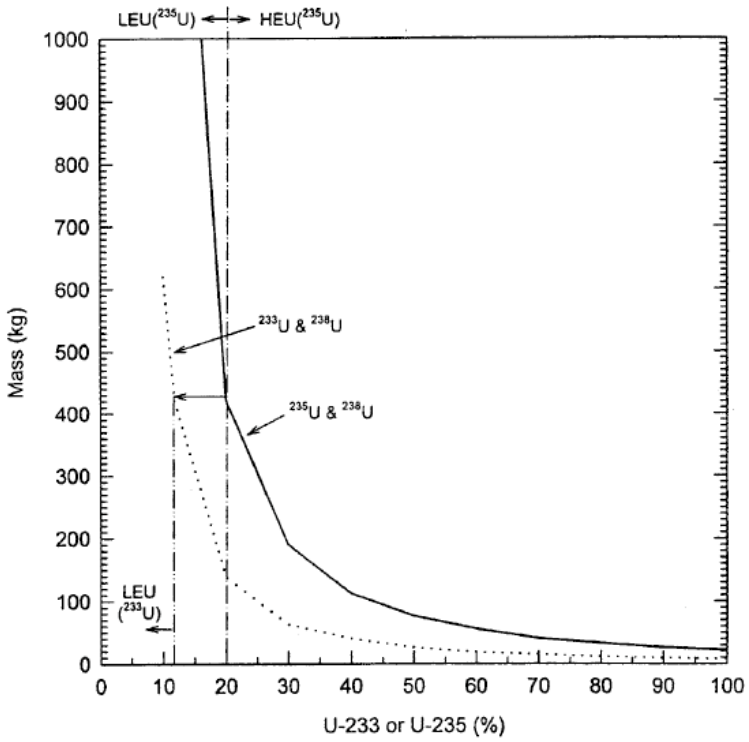

Fig. 8. Reflected critical masses as a function of percentage of U-233 or U-235 in isotopic mixture with U-238.

Table 1
Critical masses

U233/U	m_{total}, kg	m_{U233}, kg	R, crit, m
0.12	430	50	0.175
0.2	130	25	0.118
0.3	60	18	0.091
0.5	25	12	0.068
1	5	5	0.040

We can then get the number of U232 atoms in a critical mass of U and then compute the heating rate using the numbers in Table 1.

$$P = \left(heat\ rate\ in\ MeV/atom U232 \bullet year\right) \times \left(\frac{M(kgU233)}{233.04 \times 1.66054 \times 10^{-27} kg/atom}\right)\left(\frac{NU232}{NU233}\right)$$

$$\times \left(\frac{1.6021 \times 10^{-19} j/eV \times 10^{6} eV/MeV}{365.25 \times 24 \times 3600 s/y}\right) + 0.28\ W/kg \times U233/U$$

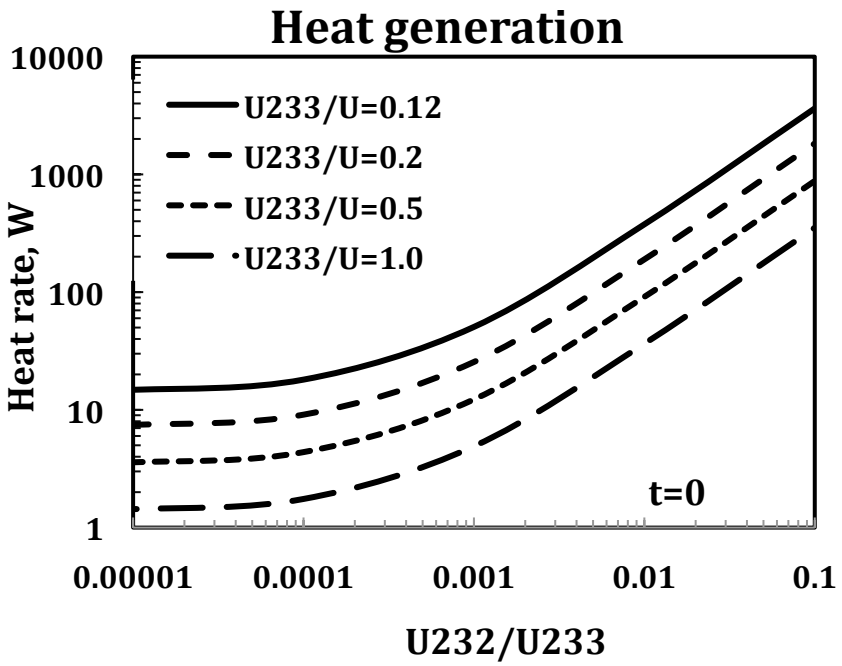

Fig. 9a. Heat rate in watts from a critical mass versus U232 concentration at time of separation of uranium.

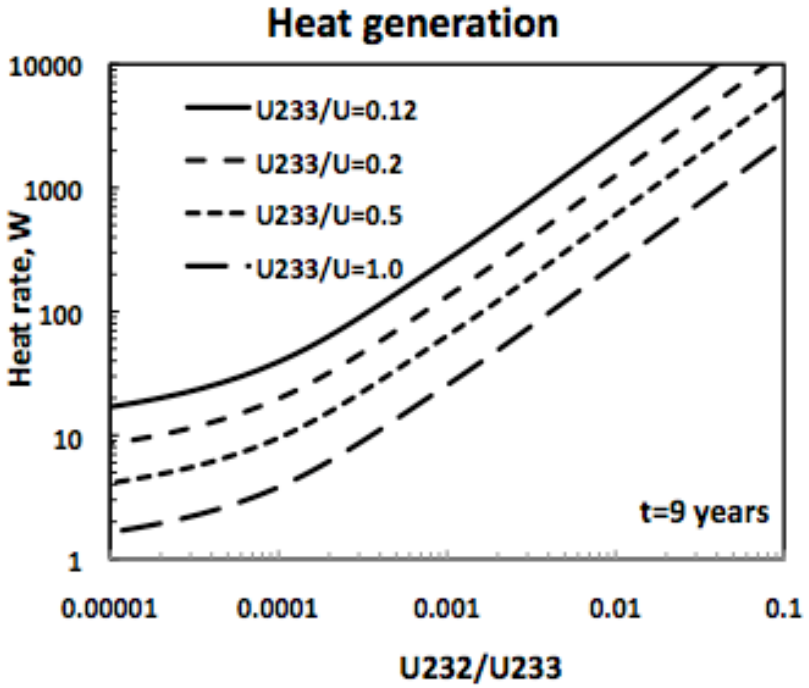

Fig. 9b. Heat rate in watts from a critical mass versus U232 concentration nine years after separation of uranium.

Notice that the heat rate is dominated by the decay of U233 until the U232/U233 ratio exceeds about 10^{-5}. At higher concentrations, the heat rate is time dependent and increases as shown in Fig. 9 by up to a factor of 40.8/5.4=7.6. The heat generation rate for U232/U233=2.4% is (641 W/kg×5 kg×0.024=77 W) 77 W immediately after

separation and increases to (5322 W/kg×5 kg×0.024=639 W) 639 W in ten years after separation. That is, 15 and 128 W/kg for these two times. It would appear the heat generation might be limiting in weapons design as will be discussed next.

EFFECTS OF HEAT GENERATION

The heat release will cause the temperature of the U232 contaminated uranium and surroundings to rise possibly causing problems if it rises too much. To get an idea of the temperature rise likely to be seen we consider here cooling of a sphere by convection in air and by radiation.

Convection heat transfer

Let us assume a surface area A and heat being generated inside by the nuclear decay process. The surface area will warm up and be cooled by convective air currents at temperature T_{air}.

$$P_{convection} = hA(T - T_{air})$$

The heat transfer coefficient, h, from the textbook of Holman (1963) and from VanSant is given below.

$$\frac{hd}{k} = 2 + 0.43(Gr_d \ \mathrm{Pr})^{1/4}, \quad 1 < Gr_d \ \mathrm{Pr} < 10^5 \qquad \text{Eq. 1}$$

The parameters describing convection in air using standard notation follows:

$$Gr_d = g\beta\Delta T d^3/\nu^2 \qquad \Delta T = T - T_{air}$$
$$\alpha = k/\rho c$$
$$\mathrm{Pr} = \nu/\alpha = 0.7 \quad in\ air\ at\ 288K$$
$$\nu = \mu/\rho$$
$$\mu = 1.78\times10^{-5}\ kg/m\cdot s \qquad in\ air$$
$$\nu = 1.46\times10^{-5}\ m^2/s \qquad in\ air$$
$$\rho = 1.23\ kg/m^3 \qquad in\ air$$
$$c = 1.005\times10^{6}\ J/kg \qquad in\ air$$
$$\alpha = 2.21\times10^{-5}\ m^2/s \qquad in\ air$$
$$\beta = 1/T$$
$$g = 9.807\ m/s^2$$
$$k = \alpha\rho c = 2.21\times10^{-5} m/s^2 \times 1.23 kg/m^3 \times 1.005\times10^{6} J/kgK = 0.273 W/mK$$
$$Gr_d P_r = 0.7\times9.807 m/s^2 \times \frac{(T - T_{air})}{T_{air}} \frac{d^3}{(1.46\times10^{-5}\ m^2/s)^2}$$

For small temperature rises of 10 K, Gr_dP_r is too large for formula of Eq. 1 to be accurate as shown below.

$$Gr_dP_r = 0.7 \times 9.807\, m/s^2 \times \frac{10K}{288K} \frac{(0.1m)^3}{(1.46 \times 10^{-5}\, m^2/s)^2} = 1.14 \times 10^6$$

For values of Gr_dP_r>10^5, the convection is turbulent and the appropriate formula from (VanSant, p 16-4, 1983) is

$$\frac{hd}{k} = 0.63(Gr_dP_r)^{1/4} \qquad \text{Eq. 2}$$

The temperature rise has been worked out for a number of examples and shown in Table 2 for a 5 kg sphere of U233 with a surface convective heat transfer to air at a radius of 0.05 and 0.5 m radius and various U232/U233 contamination ratios. The decay power appropriate to freshly separated U and after 9 years are obtained from figures 9.

Table 2
Temperature rise at surface from convective heat transfer

5 kg U233 sphere, r=0.05 m				
U232/U233	0.0005	0.003	0.024	0.1
T=0, ΔT	7 K	16 K	84 K	250 K
P	3.5 W	10 W	77 W	300 W
T=9 y, ΔT	21 K	78 K	455 K	1100 K
P	14 W	70 W	640 W	2000 W
5 kg U233 sphere, r=0.5 m				
T=0, ΔT	0.04 K	0.1 K	0.5 K	1.6 K
P	3.5 W	10 W	77 W	300 W
T=9 y, ΔT	0.1 K	0.5 K	3 K	7 K
P	14 W	70 W	640 W	2000 W

For a U233 bare sphere at 10 W heat release and 0.05 m radius the temperature is warm to the touch. Above 100 W the temperature is high and rising almost linearly with increasing power. With a sphere of radius 0.5 m surrounding the same mass of U the surface temperature rise would be small.

Radiation heat transfer

The process of heat radiation from area A to the surroundings assuming to be at temperature T_{air} is:

$$P_{rad} = \varepsilon\sigma A(T^4 - T_{air}^4) \qquad A = 4\pi r^2$$

$$T = \left(\frac{\frac{P}{\varepsilon\sigma A}}{1 - \frac{T^4}{T_{air}^4}} \right)^{1/4} = \left(\frac{P + \varepsilon\sigma A T_{air}^4}{\varepsilon\sigma A} \right)^{1/4}$$

$$\varepsilon\sigma A = 0.25 \times 5.67 \times 10^{-8} \times 4\pi r^2 \qquad T_{air} = 23\ ^{O}C$$

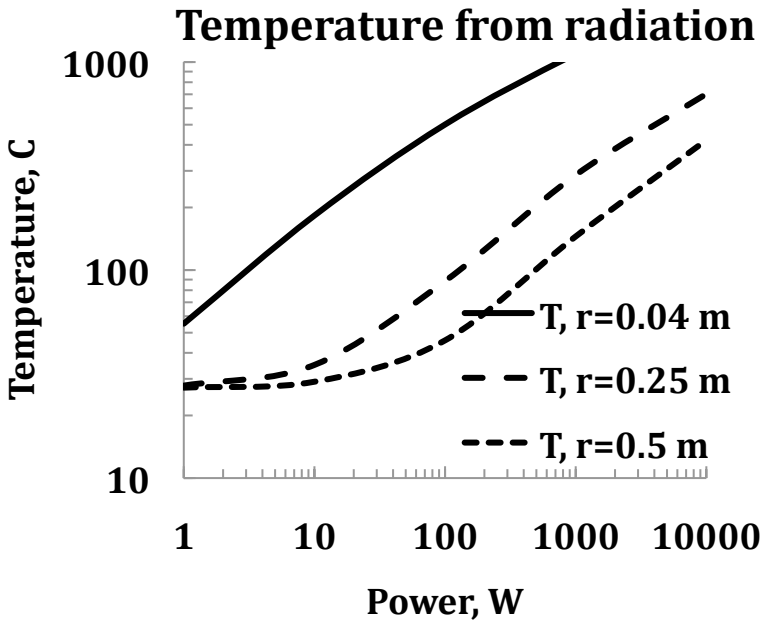

Fig. 10. Surface temperature for radiation heat transfer.

For a bare critical sphere of U233, the surface temperature will exceed 100 °C for a power generation of more than 3 W. From Fig. 9 we see that 3 W is exceeded at a U232/U233>5×10^{-4} for fresh U but after aging for 9 years it would be achieved at U232/U233>10^{-4}. If the same amount of material was radiating from a sphere of 0.25 m radius, then the temperature would exceed 100 °C for 100 W appropriate for U232/U233>0.03 for fresh material or U232/U233>0.005 for 9 year aged material. For small radius spheres convection heat transfer in air is more effective than is radiation in keeping the temperature rise limited.

DECAY POWER FOR VARIOUS MATERIALS

For general interest the decay power of various pure materials are given in Table 3.

Table 3
Decay power per kg.

Element	W/kg	Comments
Co60	17,450	GE Chart
Pa231	11.2	
Th230	0.58	
Th232	2.65E-5	t=0
Th232	2.61E-6	t=9 y, Ra228 buildup
U232	641	t=0
U232	5322	t=9 y, Th228 buildup
U235	5.8E-5	
U238	2.1E-5	
Pu238	568	
Pu239	1.9	
Pu240	7.1	
Pu241		
Pu242		
Am241	113	GE Chart
Cm242	120,000	"
Cm244	2780	"

CONCLUSIONS

The radiation associated with the thorium fuel cycle is well known and is one of the reasons it is not used in nuclear reactors, especially since hands on fabrication of solid fuel is precluded. This radiation argues against U233 from thorium use in nuclear weapons because of the dose to workers near the explosive. The allowed time of exposure is 300 hours for a fatal dose at U232/U233 =2.4%.

Not so well known is the damage to high explosive material placed near the critical mass owing to ionizing radiation. The estimated shelf life for high explosive damage is about ½ year after separation for U232/U233= 2.4%. The heat generation at the time of separation is 77 W and rises in nine years to 600 W. The temperature rise owing to this heat generation rate for a bare sphere is estimated to be 84 °C and 450 °C at time of separation and after 9 years, respectively.

References

J. P. Holman, Heat Transfer, (McGraw-Hill, New York 1963).

J. Kang and F. N. von Hippel, "U-232 and the proliferation resistance of U-233 in spent fuel," *Science & Global Security*, **9** (2001) 1-32.

L. Avrami, H. J. Jackson, M. S. Kirshenbaum, "Radiation-induced changes in explosive materials," Tech Report 4602, Picatinny Arsenal, Dover, New Jersey (1973).

J. H. VanSant, "Conduction Heat Transfer Solutions," UCRL-52863 Rev. 1 (1983).