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Spent Nuclear Fuel Project Photon Heat Deposition Calculations for Hydrogen Generation Within MCO

J. S. Lan

Westinghouse Hanford Company, Richland, WA 99352
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Abstract: Three types of water conditions are analyzed for nuclear heat deposition in a MCO: fully flooded, thick film, and thin film. These heat deposition rates within water can be used to determine gas generation during the different phases of Spent fuel removal and processing for storage.

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SPENT NUCLEAR FUEL PROJECT

**PHOTON HEAT DEPOSITION CALCULATIONS
FOR HYDROGEN GENERATION
WITHIN MULTI-CANISTER OVERPACKS**

July 1996

Westinghouse Hanford Company
P. O. Box 1970
Richland, Washington 99352

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EXECUTIVE SUMMARY

The objective of this study was to use an existing Monte Carlo geometry model of a Multi-canister Overpack (MCO) to calculate heat deposition in water, which in turn could be used to determine gas generation during the different phases of spent nuclear fuel removal and processing for storage.

Three types of water conditions (i.e., fully flooded, thick film, and thin film) are analyzed to support the baseline operational sequence, such as venting, cold vacuum drying, hot vacuum conditioning of all MCOs, and sealing during interim storage.

The MCO conservative photon source term for the heat deposition calculation has been established as a fully loaded MCO with 6.34 MTU for Mark-IV fuel. The MCO sources and the nuclear heat deposition calculational methodology are discussed in this report, but the calculation of neutron heat deposition rate is omitted due to its small source strength compared to that of the photons.

The heat deposition rates in water for full-water flooding averaged $1.17\text{E-}5$ watts per gram within the MCO, where heat deposition rates in the channels adjacent to the fuel tend to be about 30% higher than this average, and heat deposition rates in the gap at the top of the MCO are considerably less than the average. Heat deposition rates (watts/gram) for a thick water film were typically about 10% greater than the fully flooded case because the thick water film provides very little self-attenuation of the photons. The calculations predict that the heat deposition rates for very thin films of water (< 0.0001 cm thick) are about 40% less than that for the thick films, because some of the electron energy is transported out of the film.

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SPENT NUCLEAR FUEL PROJECT PHOTON HEAT DEPOSITION CALCULATIONS FOR HYDROGEN GENERATION WITHIN MULTI-CANISTER OVERPACKS

1.0 INTRODUCTION

The objective of this study was to calculate the heat deposition within the Multi-canister Overpack (MCO) that leads to the radiolysis of water, which is responsible for part of the hydrogen generation within an MCO. This analysis will support the related pressurization integration analysis (WHC 1996a and 1996b). These heat deposition rates can also be used to estimate the amount of hydrogen (H_2) and oxygen (O_2) that are liberated for potential combustible gas ($O + H$) reactions. The study includes only the analyses of the pristine MCO fuel and does not include the heat generation from sludge, oxides, or hydrides.

1.1 NEUTRON AND PHOTON SOURCE TERMS FOR MARK-IA AND MARK-IV FUEL

A detailed discussion of the use of ORIGEN2 (and RADNUC) to obtain worst case photon source terms and WIMS-E (and ORIGEN2) to obtain worst case neutron source terms for Mark-IA and Mark-IV fuel is given in the *Spent Nuclear Fuel Project FY 1996 Multi-Year Program Plan* (WHC 1996b). The source consists of 6.34 MTU of Mark-IV fuel irradiated to 16% Pu-240 (WHC 1996c). Mark-IV fuel has the higher maximum burnup, and consequently represents a worst case source for heating purposes. The photon source spectra, given in Table 1 at discrete energies, was derived from tables in *Shielding Source Term Generation for N Reactor Fuel* (Schwarz 1995). These photon sources are used in the calculations summarized in this report. Bremsstrahlung photons within the source region are also included in the ORIGEN2 calculation of the photon source.

The neutron source term in Table 2 was also derived from the same calculation, where a WIMS-E calculation was used to obtain the isotopic inventories at the end of the reactor irradiation and ORIGEN2 was used to decay these inventories for 13 years and compute the neutron production. It includes spontaneous fission, and also conservatively assumes that there is full oxidation of the fuel for the computation of the (α, n) portion of the neutron source. While this assumption slightly overestimates the total neutron source, it fully compensates for (α, n) from trace elements.

Table 1. Photon Baseline Source Term for N Reactor Fuel [decayed 13 years].

Energy (MeV)	RADNUC:ORIGEN2 Mark-IV Fuel 16.0 % ²⁴⁰Pu (photons/sec/MTU)
0.0150	2.489e+14
0.0250	5.280e+13
0.0375	5.941e+13
0.0575	5.006e+13
0.0850	2.757e+13
0.1250	2.044e+13
0.2250	2.334e+13
0.3750	1.101e+13
0.6620*	3.988e+14
0.8500	8.649e+12
1.2500	1.625e+13
1.7500	1.524e+11
2.2500	1.375e+09
2.7500	9.283e+07
3.5000	1.257e+07
5.0000	2.699e+05
7.0000	3.106e+04
11.0000	3.565e+03
TOTAL	9.173e+14

* Changed from 0.575 MeV to 0.662 MeV to accurately reflect Ba-137m gamma ray.

Table 2. Neutron Baseline Source Term for N Reactor Fuel [decayed 13 years].

Component of Source	Source Strength [Neutrons/s/MTU]
	Mark-IV Fuel
(α, n)	4.861×10^5
Spontaneous Fission	8.732×10^5
TOTAL	1.359×10^6

1.2 NUCLEAR HEAT DEPOSITION RATES

The total nuclear heat deposition within a fuel assembly would typically include: (1) the heat deposited by gamma rays that are internally produced by neutrons, (2) the heat deposited by the internal neutron source of Table 2, and (3) the heat deposited from the internal photon source of Table 1. In this particular nuclear fuel assembly, the MCO, the neutron source strength is calculated to be many orders of magnitudes less than that of the photon source shown in Tables 1 and 2. Therefore, the heat deposition from the first two items is omitted; only the last item is pursued in this report.

1.3 RADIOLYSIS OF WATER

Radiolysis results from the interaction between the radiation (α , β , γ) from fission products, transuranics, and the water environment. The energy deposited by the interaction results in the decomposition of the water. The decomposition products undergo a series of chemical reactions, after which stable reaction products are formed.

The range of an α particle in uranium is of the order of μm . The range of a β particle is slightly larger and can be expected to contribute to radiolysis locally, within a few μm of the surface. For this analysis, α radiolysis was not considered since the zirconium cladding of the N Reactor fuel element is thick enough to block all the α particle's movements.

The range of a γ ray is larger than the dimensions of an MCO. Thus, radiolysis due to γ ray attenuation is expected to be the major contributor to the pressure buildup in the MCO, through the production of hydrogen gas.

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2.0 CALCULATIONAL MODELS AND RESULTS

The widely used Monte Carlo computer code, MCNP, is employed for this calculation (Brismester 1993). MCNP has a powerful geometry routine and uses an ENDF/B database for cross sections. The quality assurance documentation of MCNP for use at the Hanford Site is given in the *Certification of MCNP Version 4A for WHC Computer Platforms* (Carter 1995). An MCNP input model typically consists of four major parts: the geometric configuration, material properties, source description, and various tallies.

The main heating source used in this calculation is the Mark-IV fuel with a reconfigured Concept 'B' geometry. A fully loaded MCO model is applied to both the flooded and the thick film calculations. For the thin film calculations, the model is reduced to a single fuel element. The plan view of the 5-tier MCNP model of the MCO for concept 'B' fuel packing is shown in Figure 1, while the corresponding elevation view is shown in Figure 2. Note that the inner hexagons shown in Figure 1 are not physically present, but are used for repeated structure modeling of a hexagonal lattice array in the MCNP computer code. For this heating source, there is a 20.3 cm (8 inch) thickness of steel shield, radially beyond the MCO, and axially below and above the fuel (Figure 2). However, this steel has been eliminated in these heat deposition calculations since we are only interested in the MCO and its internals. The MCNP input and output files are archivally saved in the Hanford common file system (CFS) in compressed tar format as shown: kbasinheat.tar.z in directory /home/jsl061.

Heat deposition tallies are made within the gaps (Figure 3) that can contain water; such as, between fuel elements (cell 15), outer water channels (cell 19), central water channels (cell 23), empty fuel slots (cell 24, Figure 1), top of MCO (cell 25, not shown), gaps above fuel (cell 26, not shown), MCO/Cask radial gap (cell 4, not shown), and bottom of the MCO, which is set to be a couple of centimeters high (no cell number, Figure 2).

The material densities and the material numbers used in the calculations are given in Table 3. Stainless steel is used for the MCO wall and the cask. Zirconium is used for fuel cladding. Fuel elements are modeled as U-238 with 0.974 wt% U-235. The corresponding cells for each material number can be found in Figures 2 and 3.

The source "geometry" consists of five tiers (baskets), placed one on top of the other inside the MCO, for a total of 270 fuel elements. The assembly end caps are conservatively ignored.

Except for the MCO-Top-Gap and the Bottom-of-Cask cases, the heat deposition at other locations are axially averaged tallies. The actual numbers vary along the height of an MCO, but not very much, because the source is constant per unit of axial length.

Figure 1. Plan View Showing Multi-canister Overpack and Fuel.

```
04/08/96 13:39:53
MCO coreg'd "B", MEIV fuel, 54
rods/level x 5 levels/MCO = 276
rods/MCO, sup&lv
probid = 04/08/96 13:37:27
basis:
( 1.000000, 0.000000, 0.000000)
( 0.000000, 1.000000, 0.000000)
origin:
( 0.00, 0.00, 200.00)
extent = ( 55.00, 55.00)
cell labels are
cell names
```

Figure 2. Elevation View Showing Multi-canister Overpack and Fuel.

```

04/08/96 13:45:47
MCO concept "B", MIV fuel 54
rods/level x 5 levels/MCO = 270
rods/MCO, inactive
probid = 04/08/96 13:37:27
basis:
( 1.000000, 0.000000, 0.000000)
( 0.000000, 0.001000, 0.999999)
origin:
( 0.00, 0.00, 200.00)
extent = ( 250.00, 250.00)
cell labels are
material numbers

```

Figure 3. Plan View of Fuel Element With Heat Deposition Tallies made in Cells 15, 19, 23, and Empty Locations Such as Cell 24.

04/08/96 13:47:40
 MCO concept "B", MKIV fuel, 54
 rods/level x 5 levels/MCO = 270
 rods/MCO, inpmkiv
 prohibid = 04/08/96 13:37:27
 basis:
 (1.000000, 0.000000, 0.000000)
 (0.000000, 1.000000, 0.000000)
 origin:
 (6.20, 3.40, 200.00)
 extent = (6.00, 6.00)
 cell labels are
 cell names

Table 3. Material Densities Used in Calculations.

Material Numbers	Materials	Densities (g/cm ³)
6	Steel	7.86000
7	Air	0.00123
1	Water	1.00000
3	Zirconium	6.55000
5	Fuel	18.77000

The calculations are made for three configurations of water, i.e., fully flooded, thick film, and thin film of water. The fully flooded case represents a pre-drained condition. The thick film case represents a post-drained stage with damp fuel surfaces, as well as 1 to 2 kg of residue water at the MCO bottom. The thin film case represents chemically bound water having gone through a cold vacuum drying process where the water thickness is down to a few Å level. The emission and propagation of the electrons and photons are described below for these water contents.

2.1 FLOODED MCO

Using a density of 18.77 g/cm³, the uranium mass for each fuel element is calculated to be 24.2 kg. There are total of 270 elements, the MCO fuel mass is 6.52 MTU with a total source strength of 5.97e15 photons per second. The average energy per source particle is 0.3449 MeV. Therefore, the total source photon energy generated by the decay of fission products is:

$$5.97e15 (\gamma/s) \times 0.3449 (\text{MeV}/\gamma) \times 1.6e-13 (\text{watts/MeV})$$

$$= 330 \text{ watts.}$$

The total heat deposited in the water of a fully flooded MCO is calculated to be 6.67 watts. Therefore, the fraction of the photon source energy deposited in the water is:

$$6.67 \text{ watts} / 330 \text{ watts}$$

$$= 2 \%$$

The total water weight in an MCO loaded with intact fuel is 571 kg. Therefore, the average gamma energy deposition within the water per gram of water is:

$$\begin{aligned} & 6.67 \text{ watts} / 5.71\text{e}5 \text{ gm} \\ & = 1.17\text{e-}5 \text{ w/g.} \end{aligned}$$

The distribution of this photon heat deposition within the water is shown in Table 4.

2.2 THICK FILM OF WATER

Similar to the flooded case, electrons generated from incoherent scattering, pair production, and photoelectric absorption in the thick film case are assumed to locally deposit all their energy.

Table 4. Photon Heat Deposition in MCO Water (Flooded).

Cells	Locations	Flooded		
		watts	grams	w/g ¹
15	Between Fuel Elements	3.20548e+00	2.51393e+05	1.27509e-05
19	Outer Channels	1.69717e+00	1.14168e+05	1.48655e-05
23	Center Channels	3.20051e-01	2.06443e+04	1.55031e-05
24	Empty Fuel Slots	9.70731e-01	1.31513e+05	7.38126e-06
25	MCO Top Gap	6.45177e-02	1.39191e+04	4.63519e-06
26	Fuel Top Gaps	4.15824e-01	3.95453e+04	1.05151e-05
4	MCO/Cask Radial Gap	5.75040e-05	2.22001e+01 ²	2.59030e-06
	MCO Total	6.67378e+00	5.71183e+05	-
	MCO Average	-	-	1.16841e-05
	Bottom of Cask ³	3.62613e-01	3.19725e+04	1.13414e-05

1. The statistical uncertainties are less than 1 %.

2. The calculation for cell 4 was made with only 5.1e-4 g/cm³ of water.

3. Portion of a number of the above cells in bottom 2 cm of cask.

For a thick film of water, one would expect less flux attenuation across the film compared to full density of water. This should translate into somewhat higher watts-per-gram numbers. An arbitrary value of $5.12000\text{e-}5$ is used as the atom density (atoms/b-cm) for the thick film of water. This is equivalent to $5.10001\text{e-}4$ gram density (g/cc) of water. The calculations of the flooded cases were repeated with this adjusted water density and the results are shown in Table 5.

It can be seen that the heat deposition rates are about 10% higher, on the average, for the thick film case compared to that of the water flooded case.

For all the calculations so far, electron transport is turned off, no electron-induced photons are created, and all electron energy is assumed to be locally deposited. Calculations have been done, which include the Bremsstrahlung photons from the electron slowing down. The comparison of heating rate calculations with or without the Bremsstrahlung photons is shown in Table 6.

The table shows that the differences are within the statistical uncertainties. This justifies the assumption that the secondary photons from Bremsstrahlung are negligible in this calculation.

Table 5. Photon Heat Deposition in MCO Water (Thick Film).

Cells	Locations	Thick Film		
		watts	grams	w/g ¹
15	Between Fuel Elements	1.78968e-03	1.28211e+02	1.39589e-05
19	Outer Channels	9.03382e-04	5.82258e+01	1.55151e-05
23	Center Channels	1.68942e-04	1.05286e+01	1.60460e-05
24	Empty Fuel Slots	6.00402e-04	6.70718e+01	8.95163e-06
25	MCO Top Gap	4.07527e-05	7.09875e+00	5.74083e-06
26	Fuel Top Gaps	2.54193e-04	2.01681e+01	1.26037e-05
4	MCO/Cask Radial Gap	8.31370e-05	2.22000e+01	3.74490e-06
	MCO Total	3.75735e-03	2.91304e+02	-
	MCO Average	-	-	1.28984e-05
	Bottom of Cask ²	2.06487e-04	1.63060e+01	1.26633e-05

1. The statistical uncertainties are less than 1 %.

2. Portion of a number of the above cells in bottom of cask.

Table 6. Bremsstrahlung Photon Contribution to Heating Rates (Watts) for Thick Film.

Cells	No Bremsstrahlung	With Bremsstrahlung
15	1.78968e-03 (0.0007) ¹	1.78991e-03 (0.0007)
19	9.03382e-04 (0.0006)	9.03641e-04 (0.0006)
23	1.68942e-04 (0.0013)	1.68867e-04 (0.0013)
24	6.00402e-04 (0.0015)	6.00280e-04 (0.0015)
25	4.07527e-05 (0.0072)	4.02726e-05 (0.0072)
26	2.54193e-04 (0.0027)	2.54081e-04 (0.0027)
4	3.36509e-06 (0.0040)	3.34911e-06 (0.0039)
Total	3.75735e-03 (0.0006)	3.75750e-03 (0.0005)

1. One standard deviation of relative statistical uncertainties.

2.3 THIN FILM OF WATER

In order to focus on the geometry right around the film, the MCO model was simplified to include only one Mark-IV fuel element; then the photon-electron transport across the outer surface of the element can be examined in detail. The assumption that all electron energy is deposited locally may no longer be valid for a thin film. Some of the electrons born in the water will escape, reducing the heat deposition. On the other hand, some electrons born in the nearby Zirconium clad may escape into the water film to increase the heat deposition.

Figure 4 shows the single Mark-IV fuel element with reflective boundary conditions at both ends of the element to represent an infinite long fuel.

The calculation here is to establish a reduction (or enhancement) factor "if" to be applied to the watts/gram for a thick film, to obtain that for a thin film. This factor f is computed by taking electron transport into account; f is defined as:

$$f = (w/g)_{\text{thin}} / (w/g)_{\text{thick}} \quad (1)$$

The modified model starts with a thick film of water of 0.5 cm thick, which should be very thick for electrons but presents little attenuation for photons. A plan view of the tally film (cell 15) is shown in Figure 4, while a vertical view of the same cell is shown in Figure 5.

Figure 4. Film of Water Plan View With Film in Cell 15.

04/08/96 13:59:33
heat deposition in thin films
within MCO, ithin3

probid = 04/08/96 13:58:40
basis:
(1.000000, 0.000000, 0.000000)
(0.000000, 1.000000, 0.000000)
origin:
(0.00, 0.00, 35.00)
extent = (4.00, 4.00)
cell labels are
cell names

Figure 5. Film of Water Elevation View With Film in Cell 15.

```

04/08/96 14:00:16
heat deposition in thin fibres
within MC0, itthin3

prodid = 04/08/96 13:58:40
basis:
( 1.000000, 0.000000, 0.000000)
( 0.000000, 0.001000, 0.999999)
origin:
( 0.00, 0.00, 35.00)
extent = ( 4.00, 4.00)
cell labels are
cell names

```

With the new model, the first step is to modify the source for one fuel element to a 1/270 of the total MCO weight. The 0.5 cm of film thickness is divided into five equal radial intervals to support a photon-only calculation. The average heat deposition rate within the 0.5 cm film is calculated to be 5.33e-6 (w/g) and the resulting radial heating profile is shown in Table 7. It shows a fairly flat distribution across the film, as expected.

The next step is to change from the photon-only mode to photon-and-electron transport mode, but using the same photon source and the film thickness as in the first step. The average energy deposition rate is calculated to be 5.28e-6 (w/g) and the resulting radial heating profile is shown in Table 8.

The ratio of the heat deposition from these two steps is nearly unity for this thick film calculation, as expected. The last step is to repeat the full electron-photon calculation for thinner films. The results are shown in Table 9.

Table 7. Radial Heating Profile for Photon-Only Mode Calculation.

	Radial Heating Profile				
Distance into Water (cm)	0.0 to 0.1	0.1 to 0.2	0.2 to 0.3	0.3 to 0.4	0.4 to 0.5
Heating (w/g)	6.15e-6	5.67e-6	5.29e-6	4.79e-6	4.69e-6

Table 8. Radial Heating Profile for Full Electron-Photon Calculation.

	Radial Heating Profile				
Distance into Water (cm)	0.0 to 0.1	0.1 to 0.2	0.2 to 0.3	0.3 to 0.4	0.4 to 0.5
Heating (w/g)	6.14e-6	5.56e-6	5.28e-6	4.96e-6	4.68e-6

Table 9. Heat Deposition at Different Film Thickness for Full Electron-Photon Calculation.

	Film Thickness Reduction				
Thin Film Thickness, cm	0.5	0.1	0.01	0.001	0.0001
Heating (w/g)	5.28e-6	5.65e-6	4.00e-6	3.05e-6	3.08e-6

At the thickness of 0.0001 cm, the energy deposition rate seems to have reached an asymptotic value. The calculations were terminated at this point. Applying the Equation 1, we determine f for a thin film as:

$$\begin{aligned} f &= (w/g)_{\text{thin}} / (w/g)_{\text{thick}} \\ &= 3.08\text{e-}6 / 5.28\text{e-}6 \\ &= 0.5784 \end{aligned}$$

Multiplying the thick-film heat deposition rates (Table 5) by this correction factor to obtain the thin film heat deposition rates, the thin film results are shown in Table 10.

Only the specific heat deposition rates (energy per unit of mass) at various locations are given in Table 10. The absolute heating rate (watts) of each location depends upon the mass of the thin film of chemically bound water after a vacuum drying process.

Table 10. Photon Heat Deposition in MCO Water (Thin Film).

Cells	Locations	Thin Film		
		watts	grams	w/g ¹
15	Between Fuels	-	-	8.07383e-06
19	Outer Channels	-	-	8.97393e-06
23	Center Channels	-	-	9.28101e-06
24	Empty Fuel Slots	-	-	5.17762e-06
25	MCO Top Gap	-	-	3.32050e-06
26	Fuel Top Gaps	-	-	7.28998e-06
4	MCO/Cask Radial Gap	-	-	2.16605e-06
	MCO Total	-	-	-
	MCO Average	-	-	7.46043e-06
	Bottom of Cask ²	-	-	7.32445e-06

1. The statistical uncertainties are less than 1 %.
2. Portion of a number of above cells.

3.0 DISCUSSION AND CONCLUSION

Although the calculations discussed so far should be considered as preliminary, the accuracy is probably sufficient considering uncertainties in 'G' values. Nevertheless, two additional accuracy concerns will be stated here.

Both concerns are in the area of thin film heating rate calculations. First, the film thicknesses listed in Table 9 should have included Ångström unit (10^{-8} cm) thickness. However, to be able to investigate the differences in such microscopic thicknesses, more computer time would have to be invested to reduce the statistical uncertainties of the Monte Carlo calculation, before any meaningful comparison can be made. It is not clear that this cost can be justified. The second concern, is the fact that the outer Zirconium clad is so thin (0.0185 cm) that there may be a need to include the electron source in the outer portion of the fuel as well. There is the possibility that the high energy electron transport from fission product decay (i.e., the Y-90), which decays with 2.282 MeV of beta particles, could increase heat deposition rates.

In conclusion, the MCO neutron heat deposition rate is omitted due to a negligible source strength. It is found that the gamma heat deposition rate in the damp case, such as in the post-draining stage, is about 10 % higher than that of the complete flooded case because of less self-attenuation within the water. For a very thin film of water, the heating rate decreases to less than 60 % that of the thick film of water, due to electrons being transported out of the film.

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4.0 UNCERTAINTIES AND ASSUMPTIONS

In general, components of uncertainties in a Monte Carlo calculation include neutron and photon sources, geometry description with the calculational models, attenuation (cross section), and statistical. In this discussion, only the statistical uncertainty in the Monte Carlo calculations, expressed as some percentage of the energy deposition rate, has been included. Because substantial detail has been included in the three-dimensional geometry calculational model, the uncertainty due to this is negligible, compared to other components of uncertainty. Similarly, photon cross sections are well known, therefore their uncertainty is also assumed to be negligible. This leaves statistical uncertainties and uncertainties due to sources. The sources for both Mark-IA and Mark-IV fuel are described in the previous section, and are obtained using conservative assumptions.

The Monte Carlo calculation gives its own estimate of the statistical uncertainty. However, there is a statistical uncertainty in this estimate of the statistical uncertainty. For uncertainties greater than 20 %, there is typically a large uncertainty in the uncertainty and the results should at most be treated as order of magnitude estimates. For uncertainties less than 10 %, the uncertainties are typically fairly reliable. For uncertainties in the 10 to 20 % range, the statistical uncertainties tend to be reasonably reliable, but should be treated with caution. The statistical uncertainties here are not attached in each table, since they are (for most of the entries) only in a range of a fraction of a percent.

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