



Monitoring Spent or Reprocessed Nuclear Fuel Using Fast Neutrons

May 15, 2011

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Over the past few years a number of experiments have been carried out at LLNL with a liquid scintillator array that has the ability to count individual MeV neutrons and γ -rays with nanosecond timing.

It has been demonstrated that this array can be used to measure the statistical properties of the neutrons emitted in individual fission chain.

The multiple time scales over which these fission neutrons are correlated allow one to deduce information regarding the nature of fissile assemblies, among which the absolute amounts of various fissile isotopes in nuclear fuels.

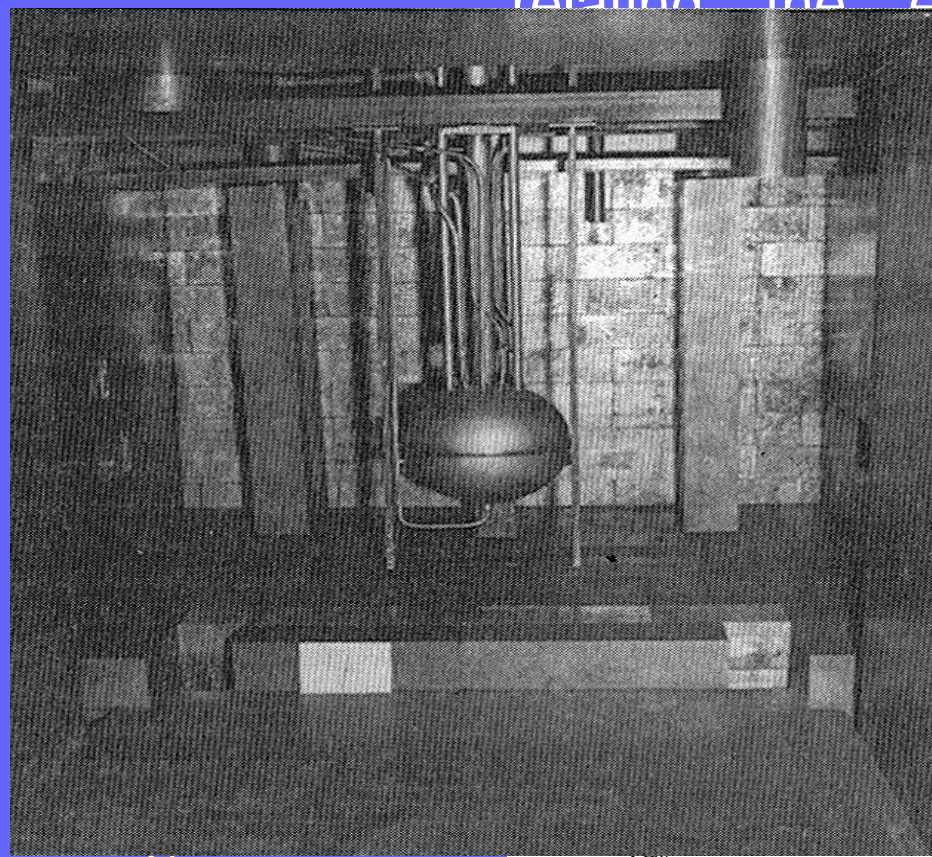
- Historical Background
- Fast neutron counting
- Assaying the Pu in MOX fuel
- Assaying the Pu in spent fuel

“Water Boiler” Reactor, Los Alamos 1944

Feynman and de Hoffman (1944) developed a formula for relating the excess neutron production relative to fission

$$\frac{\beta - \bar{c}^2}{\bar{c}} > -1$$

assembly with neutrons to the reaction M.



The theory measurements water boiler reactor carried out by E. Segre.

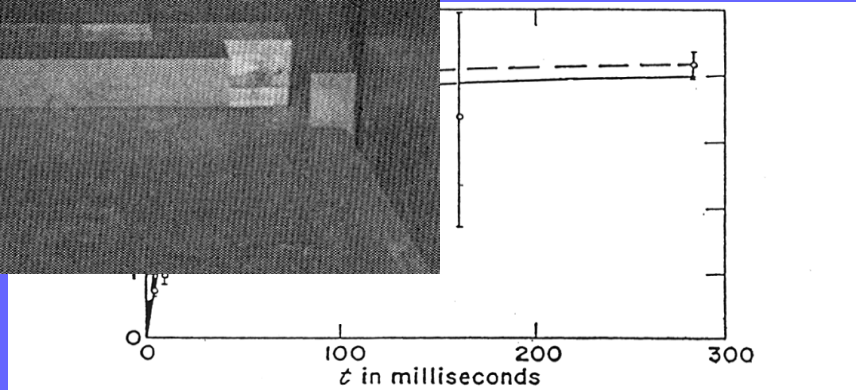
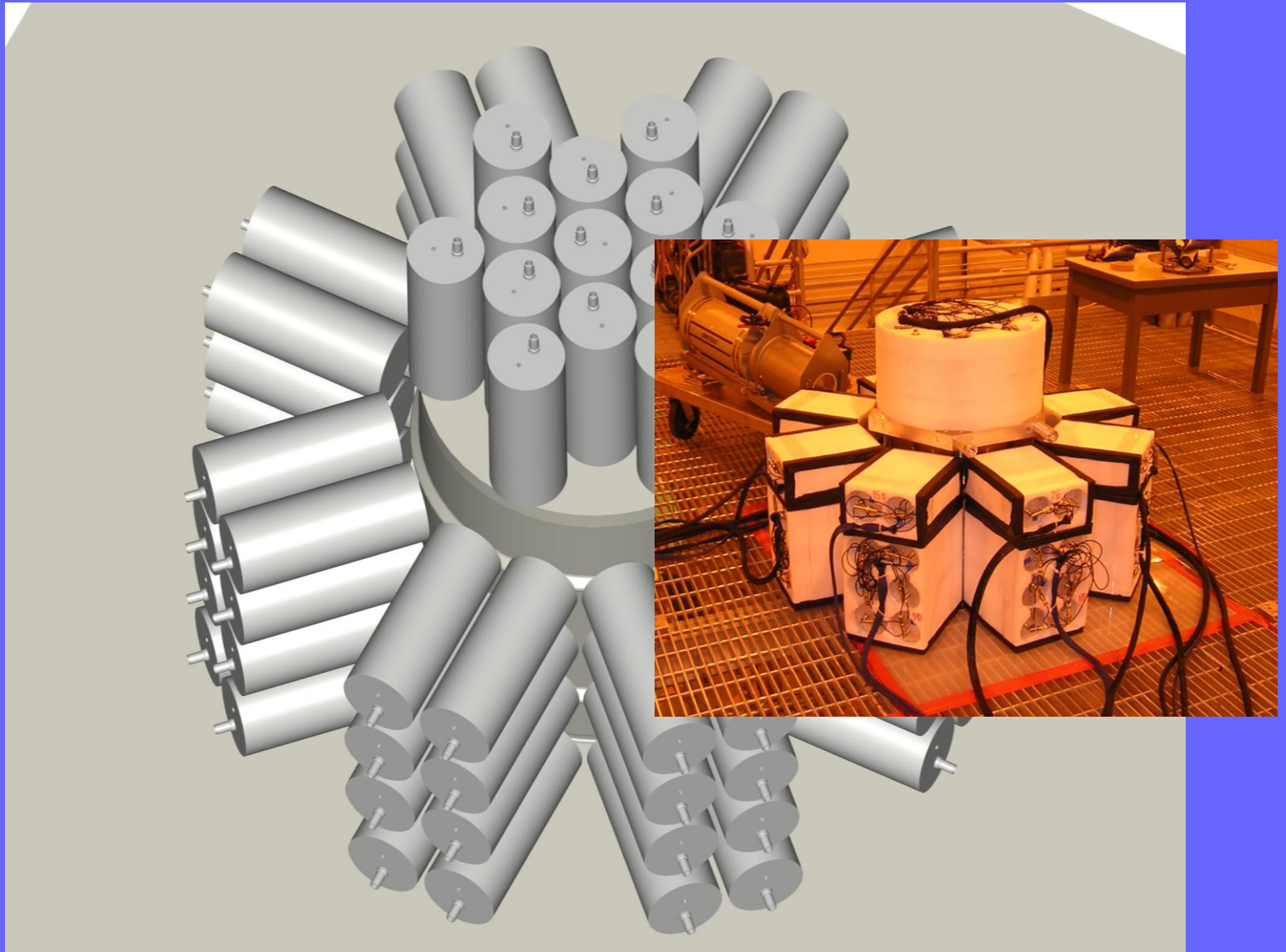


FIG. 2.—The variation of Y as a function of gate width t . All points shown were obtained by the film method except for the point at 283 msec, which was obtained by the electronic method. The dotted and solid curves were constructed as explained in Section 4.

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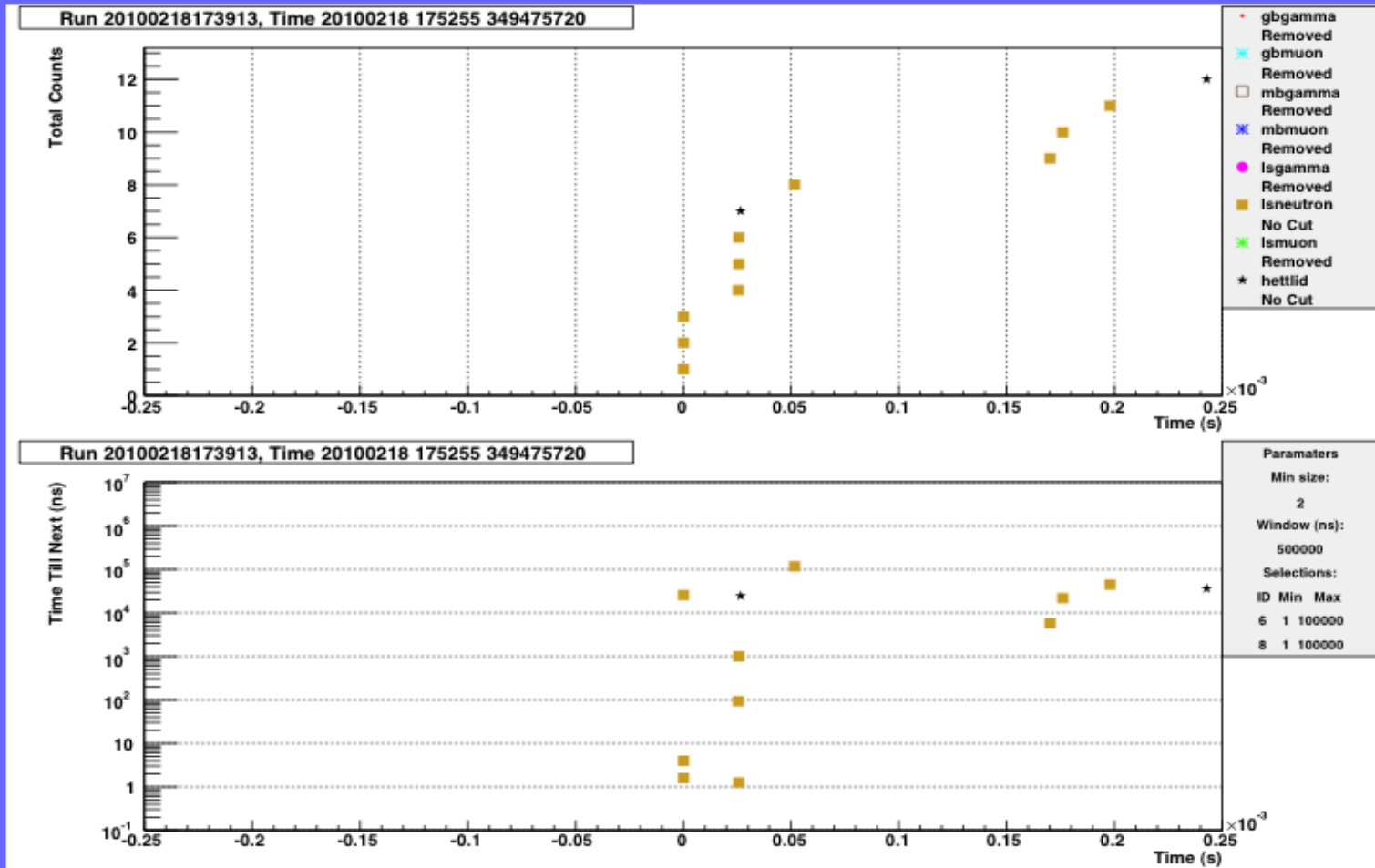
Liquid Scintillator Array





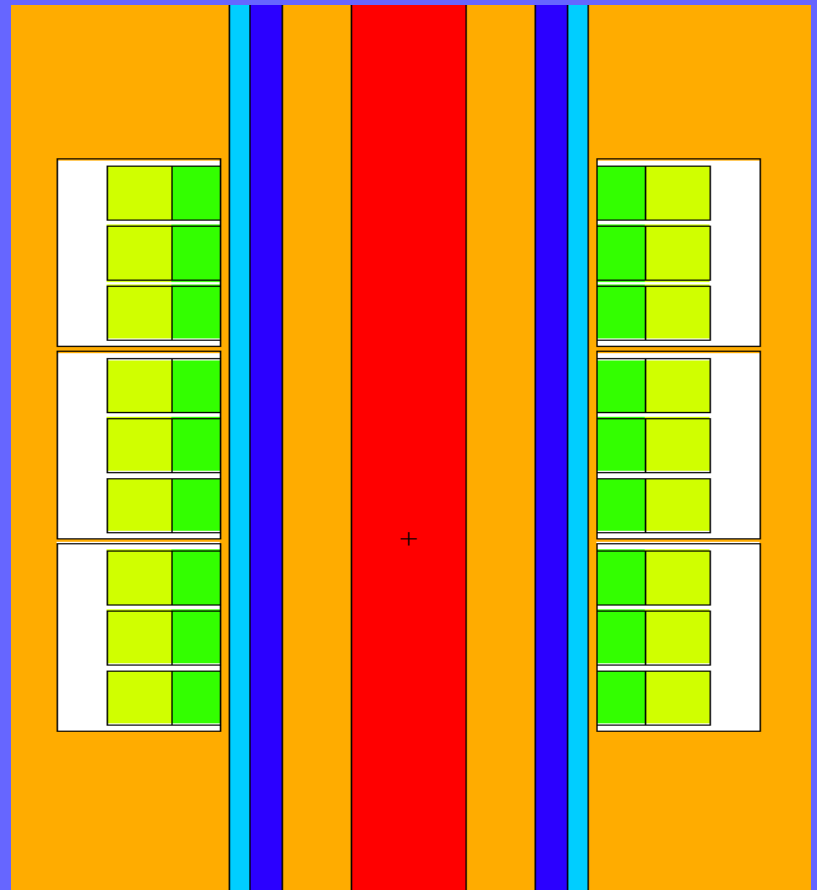
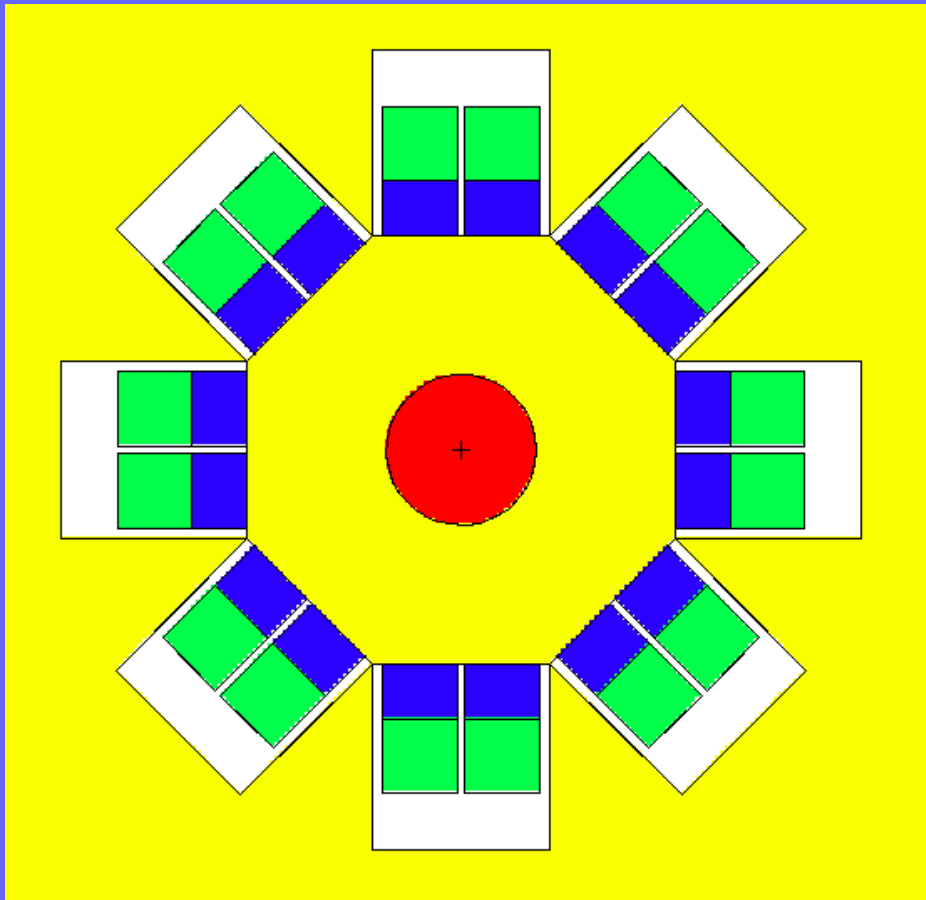
Time evolution of fission chains observed with a liquid scintillator array

822s-250microneutron

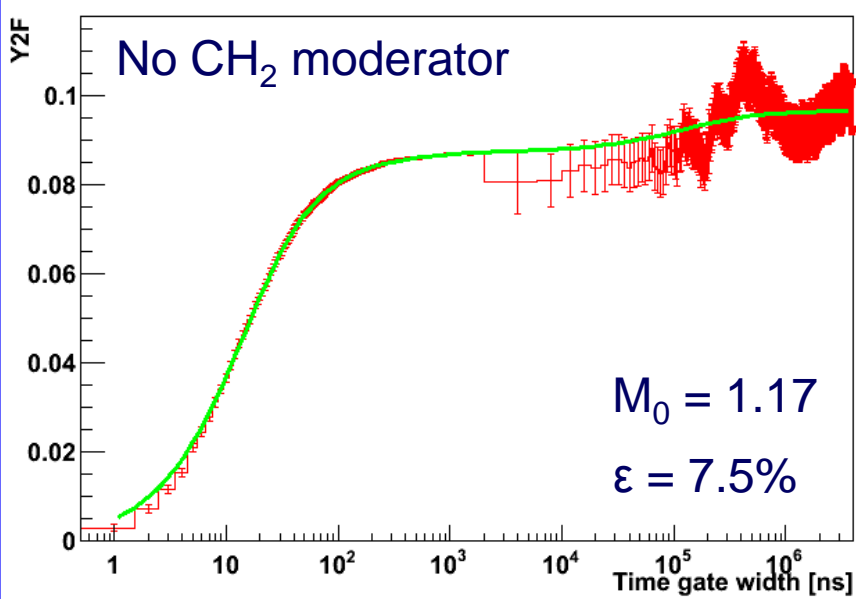


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Geometry for nuclear fuel surrounded by liquid scintillator array used in Monte Carlo simulations

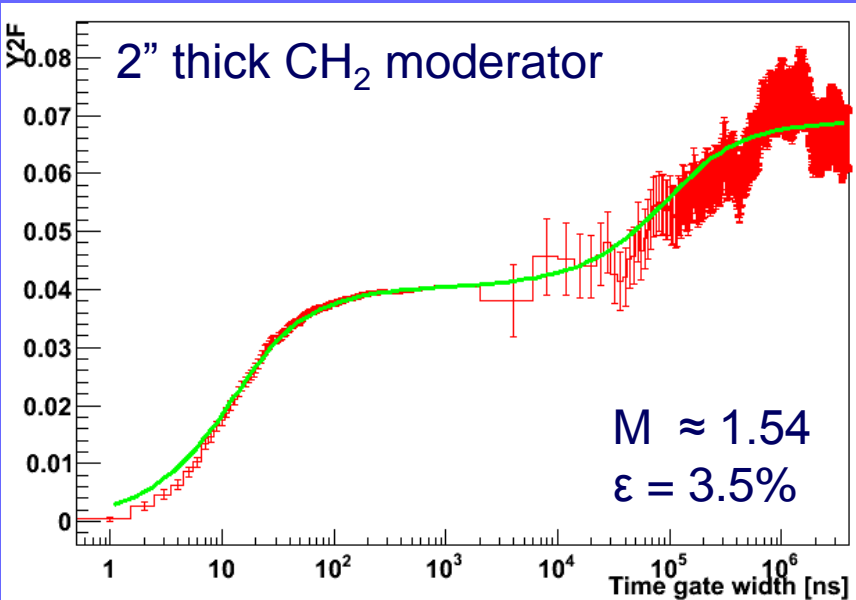


Monte Carlo Y_{2F} vs time gate for 125 kg MOX



With no moderator, fission chains happen on a timescale of tens of ns

$$Y_{2F} = \epsilon \left(M - \frac{M-1}{\bar{\nu}} \right) \left(\frac{\nu_{2s}}{\bar{\nu}_s} + (M-1) \frac{\nu_{2i}}{\bar{\nu}_i} \right) \left[1 - \frac{1 - e^{-\alpha t}}{\alpha t} \right]$$



With moderator, fission chains restart by thermal neutrons after tens of μ s

$$Y_{2F} = \epsilon \left(\frac{M_0}{M} \right)^2 \left(M - \frac{M-1}{\bar{\nu}} \right) \left(\frac{\nu_{2s}}{\bar{\nu}_s} + (M_0 - 1) \frac{\nu_{2f}}{\bar{\nu}_f} + (M - M_0) \frac{\nu_{2T}}{\bar{\nu}_T} \right) \left[1 - \frac{1 - e^{-\alpha_1 t}}{\alpha_1 t} \right]$$

$$+ \epsilon \left[1 - \left(\frac{M_0}{M} \right)^2 \right] \left(M - \frac{M-1}{\bar{\nu}} \right) \left(\frac{\nu_{2s}}{\bar{\nu}_s} + (M_0 - 1) \frac{\nu_{2f}}{\bar{\nu}_f} + (M - M_0) \frac{\nu_{2T}}{\bar{\nu}_T} \right) \left[1 - \frac{1 - e^{-\alpha_2 t}}{\alpha_2 t} \right]$$

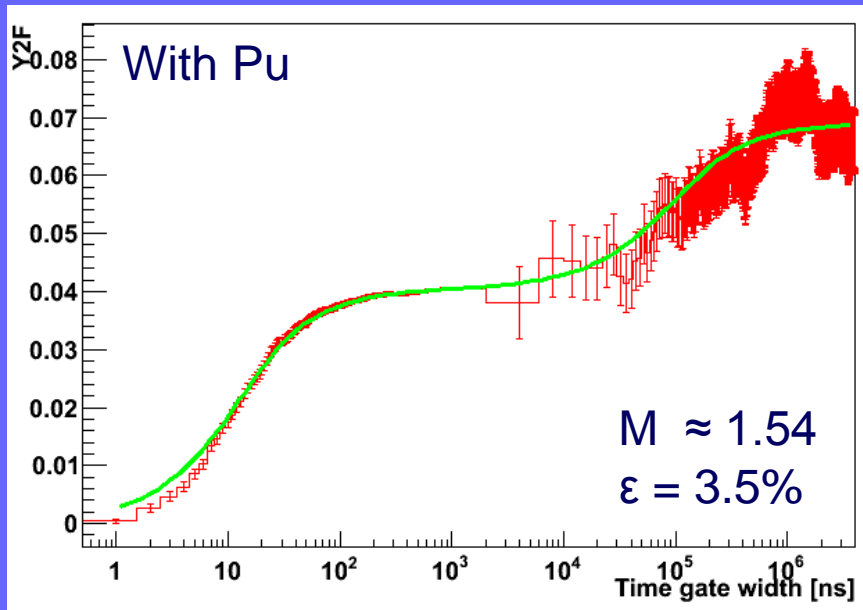


Estimating the amount of ^{240}Pu in MOX fuel

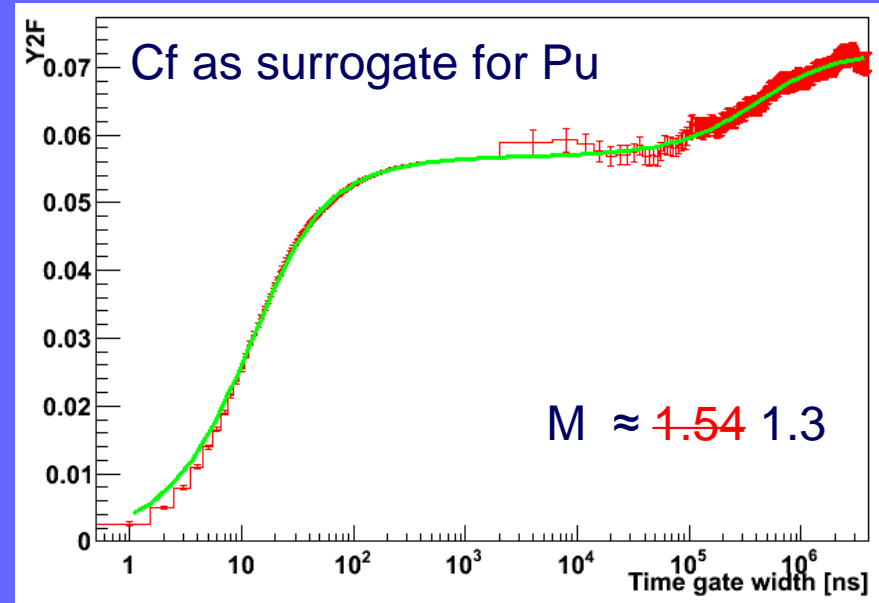
The mass of ^{240}Pu can be determined from the measured count rates, together with the inferred values of M and ϵ .

The masses inferred agree with the assumed amounts of ^{240}Pu in our simulated MOX fuel.

Diversion of Pu for proliferation



Chemical diversion of Pu for proliferation can be detected even if surrogate ^{252}Cf is used to mask count rate differences.



Estimating the total amount of Pu in MOX fuel

The amount of $^{239}\text{Pu} + ^{241}\text{Pu}$ in MOX nuclear fuel can be inferred from the asymptotic value for Y_{2F} when there is an external CH_2 neutron moderator:

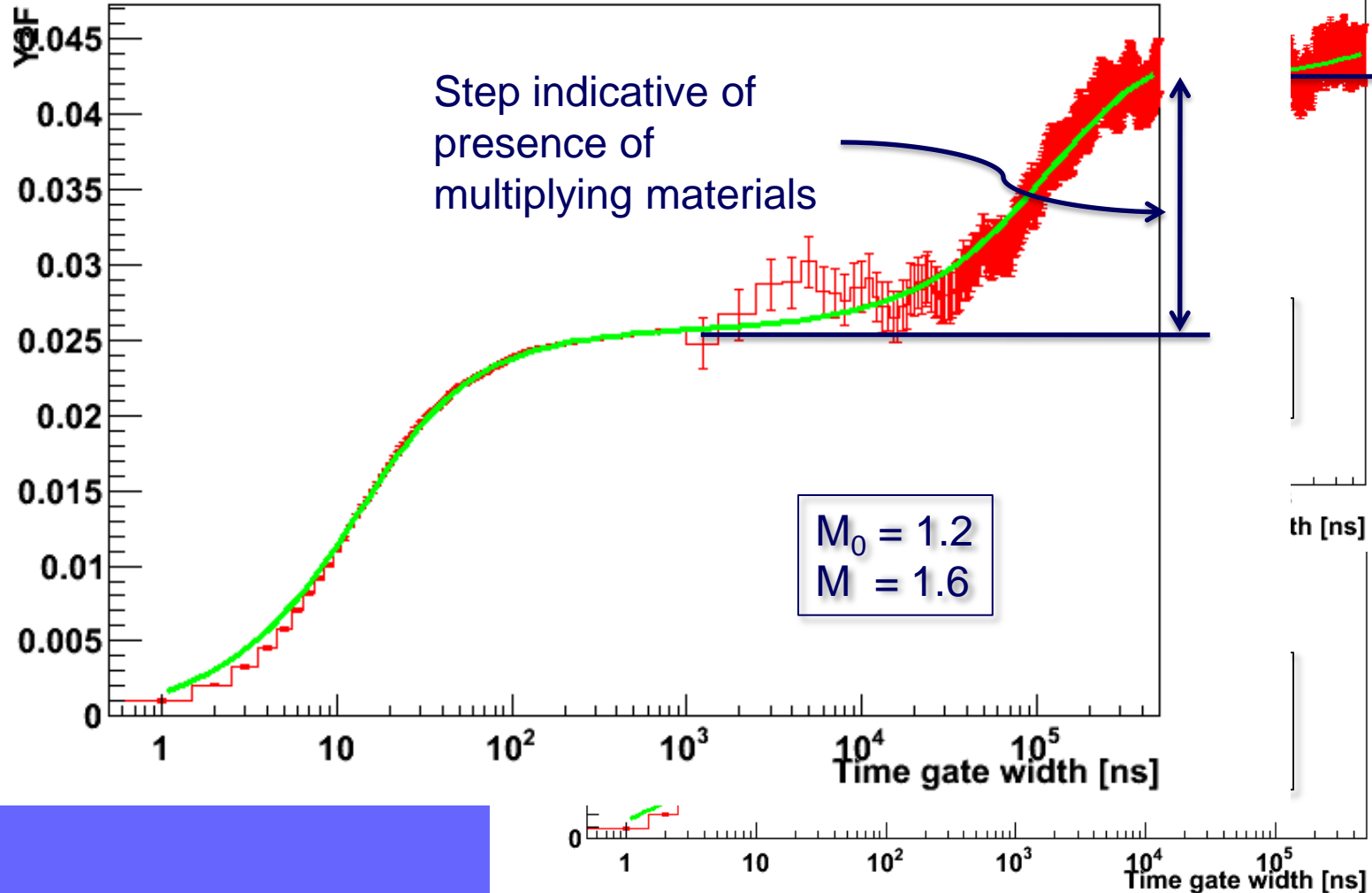
$$Y_{2F} \rightarrow \varepsilon \left(M - \frac{M-1}{\bar{\nu}} \right) \left(\frac{\nu_{2s}}{\bar{\nu}_s} + (M_0 - 1) \frac{\nu_{2f}}{\bar{\nu}_f} + (M - M_0) \frac{\nu_{2T}}{\bar{\nu}_T} \right)$$

The excess of M over M_0 is due to the contribution of fissions in ^{239}Pu , ^{241}Pu , and ^{235}U induced by thermalized neutrons.

The value for M we obtain is consistent with the amounts of these isotopes used in the simulations.

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Moderation time scale Y_{2F} for spent fuel shielded with 2" CH_2 and 4" Pb





Can we infer the amount of Pu + ^{235}U ?

By adding a 2" thick CH_2 layer around the spent fuel, we can measure the increased multiplication, that is the fraction of induced fissions due to ^{239}Pu + ^{241}Pu + ^{235}U .

Monte Carlo calculations can help to predict the number of thermally induced fissions as a function of the mass of these isotopes.

Comparison of the number of induced fissions with and without CH_2 may allow us to distinguish the amounts of ^{235}U versus ^{239}Pu + ^{241}Pu .



Conclusions

- Our Monte Carlo simulation results suggest that fast neutron counting techniques ought to be very useful for assaying the amount of Pu in reprocessed nuclear fuel
 - The short time behavior of Y_{2F} and Y_{3F} can be used to assay the amount of ^{240}Pu in the reprocessed fuel.
 - If the reprocessed fuel is surrounded with a few inches of moderator, the long time behavior of Y_{2F} can be used to determine the absolute amount of $^{239}\text{Pu} + ^{241}\text{Pu}$ in the reprocessed fuel.
- For spent fuel the neutron counting statistics are marginal due to the need to use a high Z shield thick enough to block the enormous γ -ray flux. However, the statistics may be good enough to use Y_{2F} to at least estimate the amount of $^{239}\text{Pu} + ^{241}\text{Pu} + ^{235}\text{U}$ in spent fuel. We are still looking into whether we can identify the amount of Pu.

Questions ?

