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ABSTRACT

A thermal neutron detector has been developed that integrates the neutron capture medium (^{6}LiF) and scintillator (ZnS) into a thin screen, which is coupled to wavelength shifting fiber-optic ribbon. The ^{6}LiF and ZnS powders are incorporated in a hydrogenous binder. The detector is constructed of alternating layers of the capture/scintillator screen and the fiber-optic ribbon. The scintillation light produced in the ZnS is absorbed and reemitted in the fibers and is transported to photomultiplier tubes (PMTs).

The detector is sensitive to gamma radiation, but the difference in the pulse decay times of gamma-ray and neutron events allows for discrimination using pulse shape analysis (PSA). To determine the feasibility of using PSA with this detector, a number of intrinsic characteristics of the detector have been measured; specifically, the number of photoelectrons produced at the photocathode of the PMT from the average neutron capture event in the screen and the temperature stability of the detector with respect to pulse shape.

The number of photons that reach the PMTs was measured with two different PMTs. One was a typical PMT with single-photoelectron resolution, and the other was a Hybrid PMT comprised of a photocathode coupled to a PIN diode with few-photoelectron resolution. The number of photons that reach the PMTs is between 1600 and 2200, which is sufficient for PSA. The sensitivity of pulse shape to temperature has also been evaluated. Although the pulse decay time changes with varying temperature between –25 to +50 °C, the normalized detector pulses have approximately the same amplitude around 400 ns. This results in a stable zero-crossing time of the 400-ns delay-line shaped pulse, and thus the PSA is independent of temperature.

A four-sided prototype well counter has been built. It has a short neutron die-away time ($\tau < 5$ μs), which increases its sensitivity to measurements of ^{240}Pu by neutron coincidence counting. Because of the high α,n-neutron yields in pyrochemical residues, greater sensitivity is required to measure these materials. Counters with relatively long τ (for example, those that use ^{3}He detectors) will suffer from high accidental coincidence rate. Results with the prototype counter from high-rate measurements, equivalent to those expected from residues, are presented.

INTRODUCTION

Neutron coincidence counters (NCC) have been valuable tools for the nondestructive analysis (NDA) of special nuclear materials. These counters utilize the correlated fission neutrons emitted from a plutonium-containing sample to determine the mass of plutonium present. A new NCC using a scintillator coupled to a fiber-optic ribbon has been developed to address measurement limitations inherent in counters utilizing ^{3}He detectors. The counter is shown in Fig. 1. The neutron coincidence counting technique is based on the principle that the fission of a plutonium nucleus can result in two or more neutrons being liberated nearly simultaneously. The four-sided, scintillator-based well counter [1, 2] was designed to have a short die-away time ($\tau < 5$ μs). It also exhibits good neutron detection efficiency (ε ~30%). By comparison, NCCs designed with ^{3}He tubes and polyethylene-moderator typically have die-away times of 22–50 μs and efficiencies of 40%–65%. The die-away time dictates the coincidence gate width, and a short die-away time reduces the number of accidental coincidences. The "accidentals" drive the measurement precision of neutron coincidence counting to unacceptable levels when (α, n) neutron yields are large and die-away times are long. The primary utility of such a detector is the accurate measurement of plutonium, specifically ^{240}Pu, by neutron coincidence counting in materials with high-uncorrelated (α, n) yields. The target materials would be salt residues that contain ^{240}Pu from pyrochemical processes.

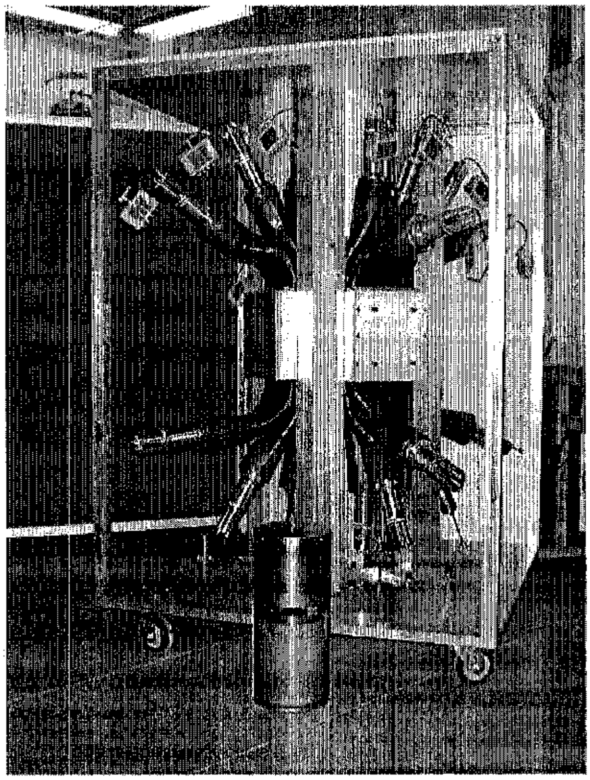

Fig. 1. Shown above is the prototype scintillator based well counter. One can clearly see two of the four panels with the associated hardware. The sample holder is shown in the foreground, which is inserted into the middle of the counter via a hole in the top.

Because of the high intrinsic gamma-ray yield associated with nuclear fission and actinide decay processes, the gamma-ray flux will typically exceed the neutron flux through the detector. For accurate measurements, the detector is required to be either insensitive to gamma rays or have the ability to distinguish gamma-ray interactions from neutron events. The ^{6}LiF/ZnS (Ag) scintillator and fiber-optic system benefits from a higher density of neutron capture nuclei (^{6}Li) relative to ^{3}He in gas tubes, and the homogeneous mixture of neutron moderator (hydrogenous binder in the screen and the polystyrene fibers) and capture medium. The resulting shorter die-away time reduces the accidental coincidence rate and permits measurements of materials with high yields of uncorrelated (α, n) neutrons. One drawback of a scintillator system is that it is sensitive to gamma rays. However, scintillators based on ^{6}LiF/ZnS (Ag) have the characteristic that pulse decay times (shapes) depend on the charge density in the scintillating ZnS (Ag) crystal. The charge density is lower for Compton electrons from gamma-ray interactions compared to energetic tritons and alpha particles produced by neutron capture. Thus, pulse-shape analysis (PSA) is a method that is used in neutron counting with scintillators to discriminate against gamma rays. Typical preamplifier pulses from gamma-ray and neutron interactions are shown in Fig. 2. Custom PSA electronics has been developed using a combination of pre-filtering and delay-line shaping coupled with a modified time pickoff.

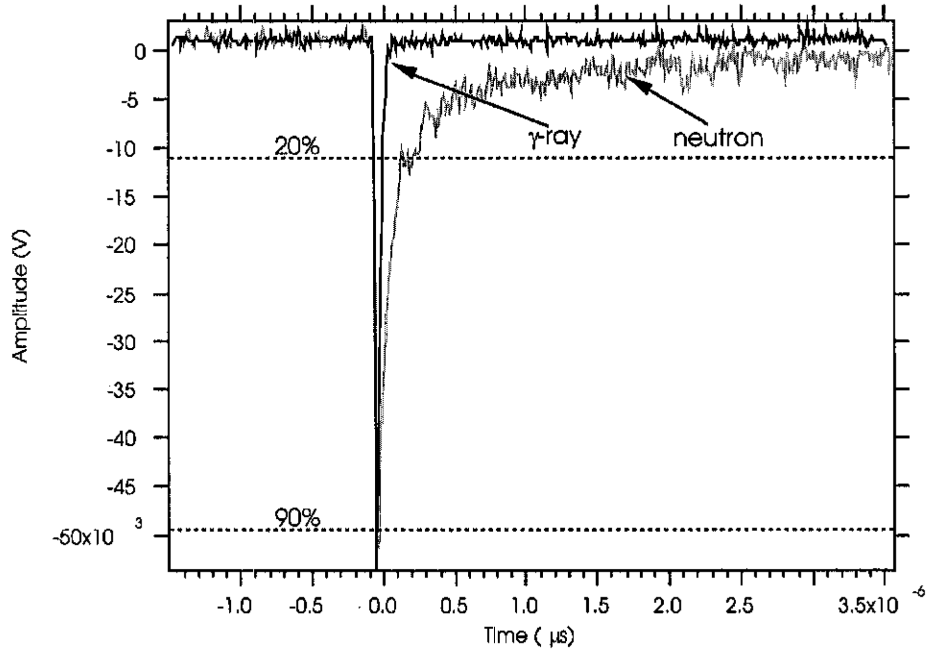

Fig. 2. Typical pulses acquired from gamma-ray and neutron events in the detector. It is evident that the neutron pulse has a much longer decay time compared to the gamma-ray pulse.

MEASURING THE AVERAGE NUMBER OF PHOTOELECTRONS

The statistics, i.e., the amount of charge represented by the PMT pulse, determines the magnitude of the high-frequency fluctuations that cause the long-tail pulse from neutron events to cross zero and be counted as new events. Smoothing of the fluctuations can be done with electronic filtering at the expense of timing of the shaped pulse. Knowledge of the average number of photoelectrons produced at the photocathode per neutron capture event is important to design for optimized filtering versus timing and optimized lower-level discriminator setting versus detection efficiency.

The details of this measurement have been reported [3] with a smaller prototype detector. The "small slab" consists of 20 layers of scintillator and fiber ribbon. The active area of the screens is 10 × 15 cm. The average number of photoelectrons produced per neutron capture event in the slab was independently measured with two PMTs that had single and few-photoelectron resolution. The ETL 9921QB[A] PMT with single-photoelectron resolution and the DEP PP0275C[B] hybrid PMT with few-photoelectron resolution were used. The neutron source used was a ^{252}Cf source, inside a tungsten shield and surrounded by polyethylene for moderation.

From two unique photomultiplier tubes it was determined that the number of photoelectrons produced for the average neutron event in the detector was between 130 and 174. Assuming a quantum efficiency of 16% for each PMT, the average number of photons incident on the photocathode is between 812 and 1087. Normal operation of the detector will have a PMT coupled to each end of the fiber bundle resulting in ~ 1600 to 2200 photons arriving at the photocathode of the PMTs.

TEMPERATURE STABILITY

Timing for reliable PSA requires that the pulse shape from the detector be independent of temperature. Because photomultiplier tubes and acquisition electronics are relatively stable with respect to temperature, the BC-704 neutron-capture/scintillator screens were tested for temperature stability [4]. Bare screens of BC-704 were coupled to a Burle S83049F photomultiplier tube and placed in a well-regulated environmental chamber. Pulses from the preamplifier were digitized and averaged to give a representative pulse shape at four temperatures. Those pulse shapes are shown in Fig. 3. The inset plot in Fig. 3 shows that at ~400 ns the pulse amplitude normalized to the peak amplitude is nearly the same regardless of temperature.

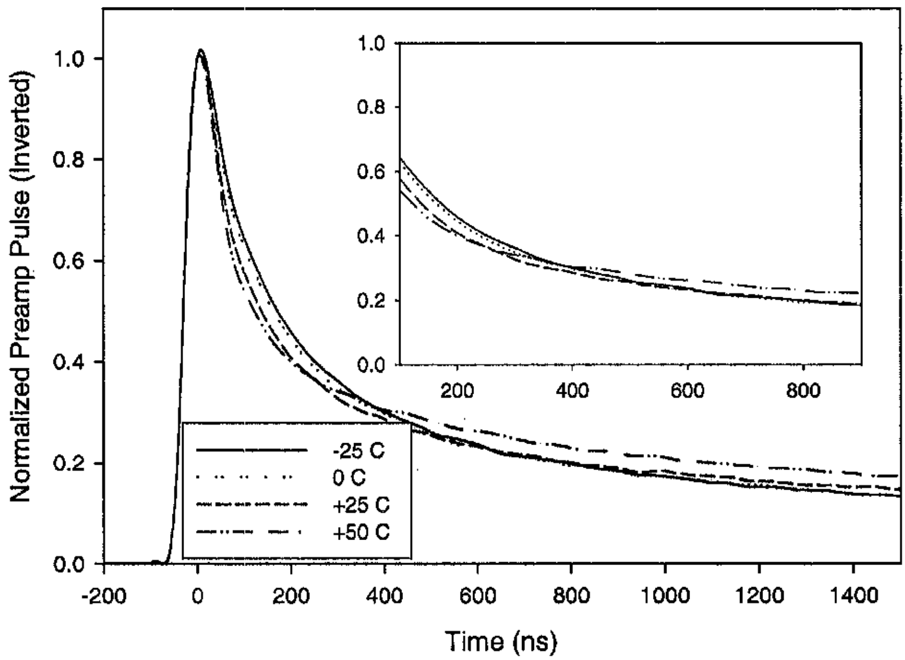

Fig. 3. Preamplifier pulses at various temperatures. The inset figure shows the stability of the normalized pulses near 400 ns.

Because of this pulse stability near 400 ns with respect to temperature, 400 ns delay-line shaping was used to generate the pulses used for PSA timing. These delay-line shaped pulses are shown in Fig. 4 at –25 °C and +50 °C and exhibit a very stable zero crossing near 400 ns with respect to temperature.

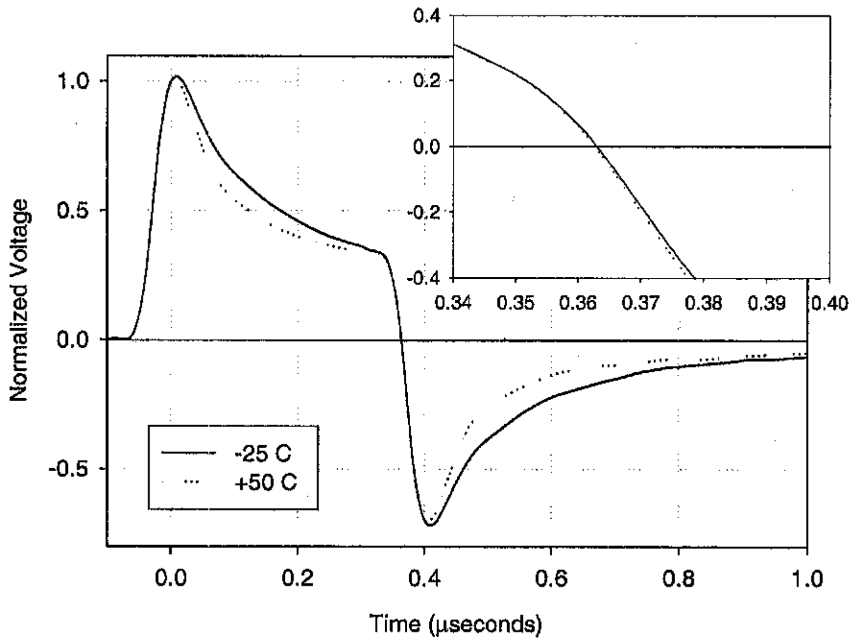

Fig. 4. Single delay-line shaped pulse as a function of time at –25 °C and 50 °C. The inset figure shows the consistent zero crossing of the shaped pulse near 400 ns regardless of temperature.

HIGH-RATE TESTS

This prototype well counter was designed to measure pyrochemical residues from separation processes. The materials typically contain plutonium and americium in oxide form and salt residue. Some residues come from extractions of americium from multiple metal feed batches into a salt, or from such a residue that is "scrubbed" to remove plutonium from the molten salt. The combination of alpha-emitting actinides and low-Z materials will result in large yields of uncorrelated (α, n) neutrons. The ratio of the yields of (α, n) to spontaneous fission neutrons is represented by the symbol α. The accuracy of various counters as a function of α is shown in Fig. 5. The Thermal Neutron Multiplicity Counter (TNMC) [5] is a counter based on 4-atmosphere ^{3}He tubes and has a τ of approximately 50 μs and ε of about 40%. The Epithermal Neutron Multiplicity Counter (ENMC) [6] is a counter based on 10-atmosphere ^{3}He tubes to achieve a τ of 22 μs and an ε of 65%.

The Fiber counter is the well counter based on ^{6}LiF/ZnS (Ag) screens and wavelength-shifting fibers that is the subject of this paper. An optimized version of this counter would have a τ of approximately 4 μs and ε of 50%. Fig. 5 clearly shows that the measurement uncertainty increases with increasing α, so in order to obtain reasonable uncertainty, counters with small die-away times and high efficiency are necessary to count those materials with high α. The data in Fig. 5 shows a comparison of the relative standard deviation in the measured plutonium mass for a 60-g plutonium sample as a function of α. The relative standard deviation for each counter was calculated from a figure-of-merit code [7].

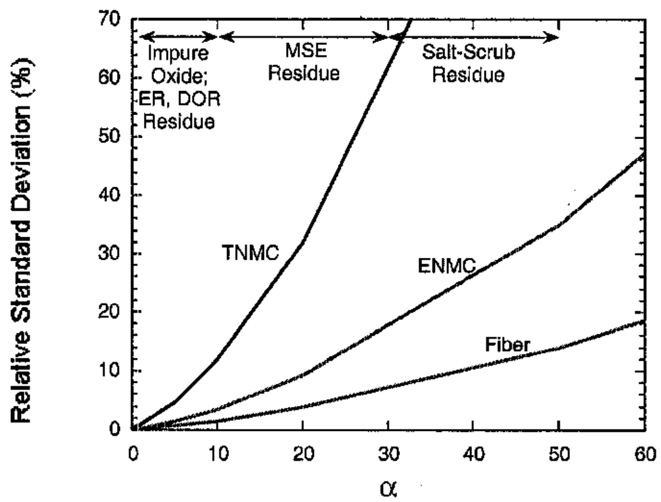

Fig. 5. Relative standard deviation as a function of α for three neutron coincidence counters. This graph shows that to measure plutonium in high α residues a detector with high efficiency and short die away time is desired to obtain smaller uncertainties.

A worst-case example of a pyrochemical residue might contain 600 g of weapons grade plutonium in oxide form and an α value of 50. In addition, 680 g of ^{241}Am (also in oxide form) is required to attain α = 50 with this quantity of plutonium containing 6% ^{240}Pu. The total neutron rate from such an extreme residue is 1.84 million neutrons per second. This rate is distributed among the 12 PSA channels for the normally configured counter.

A set of high-rate tests of the individual channels of the counter determined that the counter can measure 600 g of plutonium in a residue with α = 50. A ^{252}Cf source simulated the total neutron rate of the residue described above. Its total neutron yield was only ~350,000 neutrons per second at the time of the test. However, the outputs of six detector elements (two full sides of the four-sided counter) were combined into one electronics channel (one of the custom PSA electronics boards) to boost the effective neutron yield of the source six-fold to 2.10 million, which exceeds the theoretical rate of the example residue.

Several sets of PSA data were obtained in the same high-rate test at varying event thresholds. Some of the PSA (time) spectra are shown in Fig. 6. As the event threshold was lowered, more gamma-ray events (first peak) are processed compared to neutrons (second peak). The events contained in the region between the gamma-ray and neutron peaks are neutron/gamma-ray pileup events. Their neutron/gamma-ray pileup origin was verified by taking PSA spectra with a pure gamma-ray source (^{137}Cs) that gave a count rate that matched the total event rate (neutrons plus gammas) of the ^{252}Cf source at a 100-mV event threshold. The ^{137}Cs and ^{252}Cf spectra are shown in Fig. 7 at the 150 mV event threshold. It is clear from Fig. 7 that few gamma-ray events are contained under the neutron peak. Five regions of interest (ROI) were set in the ^{137}Cs and ^{252}Cf PSA spectra as shown in Fig. 6. The integrated counts in each ROI from the ^{137}Cs source as a function of event threshold were normalized to the corresponding ROI counts obtained with the ^{252}Cf source; these ratios are shown in Fig. 8 as a function of event threshold. At a threshold of 150 mV, less than 15% of the events in ROIs 2 and 3 in the ^{252}Cf spectrum show up in the ^{137}Cs spectrum. Additionally, for ROIs 4 and 5, less than 1.6% of the events in the ^{252}Cf spectrum show up in the ^{137}Cs spectrum. This indicates that the events in ROIs 2–5 are primarily due to neutron and neutron/gamma-ray pile-up events.

The neutron PSA gate for coincidence measurements will be set to include events that would register in ROIs 2,3,4, and 5 of the PSA spectrum. The neutron detection efficiency of the counter as a function of event threshold is shown in Fig. 9 and will be 23% at the 150-mV threshold setting.

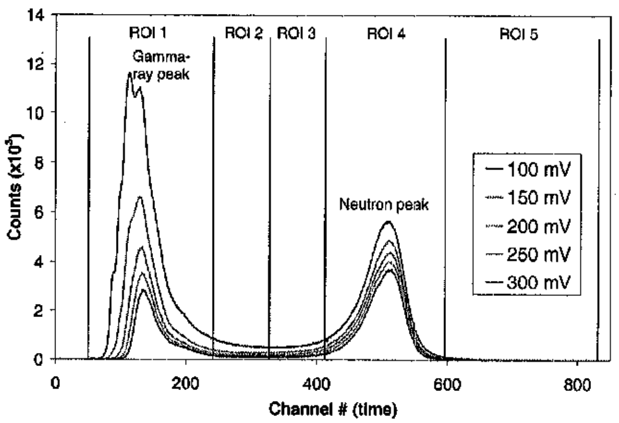

Fig. 6. PSA (time) spectrum as a function of event threshold.

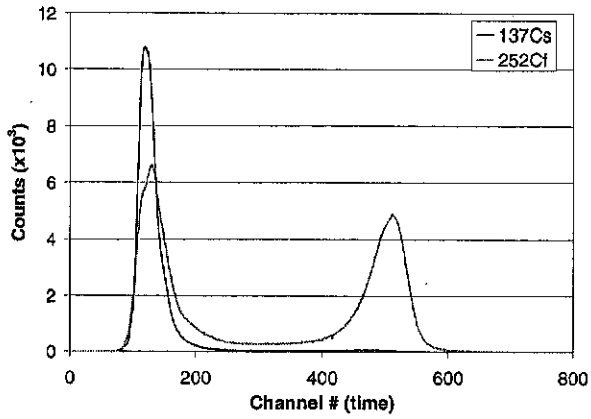

Fig. 7. PSA spectra with gamma-ray source (^{137}Cs) and gamma-ray and neutron source (^{252}Cf).

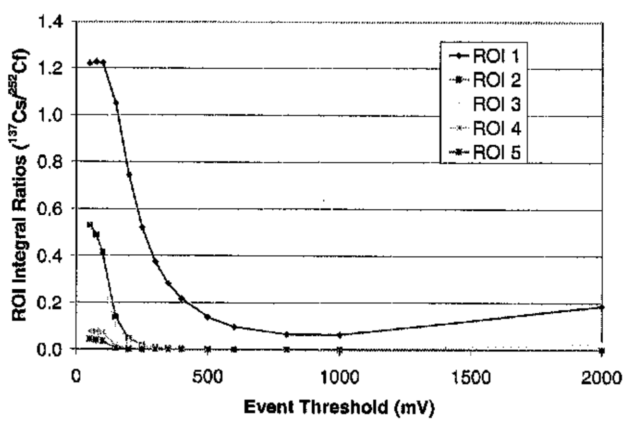

Fig. 8. Ratio of integrated ^{137}Cs ROI counts to ^{252}Cf ROI counts.

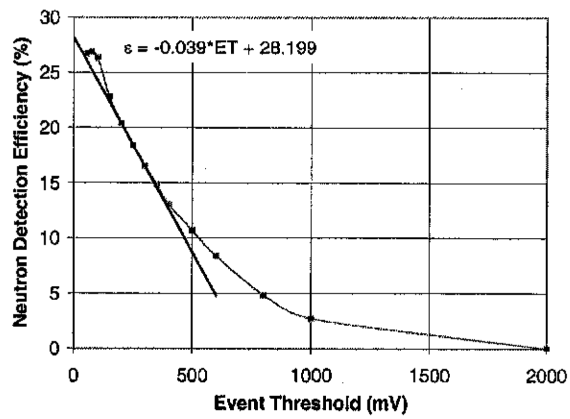

Fig. 9. The neutron detection efficiency as a function of event threshold. The extrapolated line shows a "zero-threshold" efficiency of ~28%.

The "zero-threshold" efficiency of 28% agrees well with the intrinsic efficiency of 30% calculated using MCNP.

CONCLUSIONS

The PSA of the "as built" well counter described in this paper was successful (for gamma-ray discrimination) at rates equivalent to 600 g of weapons grade plutonium in high-α,n (α=50) residues. The neutron detection efficiency of the counter optimized for measurements of such residues at high rates will be ~23% depending on the event threshold. The stability of the PSA was demonstrated over a range of –25 °C to +50 °C. Additional measurements show that changes in neutron detection efficiency for temperature changes of ±10 °C are within ±2%.

FUTURE WORK

Coincidence measurements of ^{252}Cf sources and plutonium oxide samples in various matrices will begin in the fourth quarter of FY2001. Advanced gamma-ray discrimination techniques without delay-line pulse shaping will be tested as well.

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