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Instrumentation and Controls Division

**DESIGN OF A CALIFORNIUM SOURCE-DRIVEN MEASUREMENT SYSTEM FOR
ACCOUNTABILITY OF MATERIAL RECOVERED FROM THE MOLTEN SALT
REACTOR EXPERIMENT CHARCOAL BED***

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**Design of a Californium Source-Driven Measurement System for Accountability of
Material Recovered From the Molten Salt Reactor Experiment Charcoal Bed**

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ABSTRACT

The Molten Salt Reactor Experiment Facility (MSRE) operated from 1965 to 1969. The fuel was a molten salt that flowed through the reactor core which consisted of uranium tetrafluoride with molten lithium and beryllium salt used as the coolant. In 1968 the fuel was switched from ^{235}U to ^{233}U . The Molten Salt Reactor Experiment was canceled in 1969 at which time approximately 4800 kg of salt was transferred to the fuel drain tanks. There was about 36.3 kg of uranium, 675 grams of plutonium and various fission products present in the fuel salt.(3) The salt was allowed to solidify in the fuel drain tanks. The salt was heated on a yearly basis to recombine the fluorine gas with the uranium salt mixture. In March 1994, a gas sample was taken from the off gas system that indicated ^{233}U had migrated from the fuel drain tank system to the off gas system. It was found that approximately 2.6 kg of uranium had migrated to the Auxiliary Charcoal Bed (ACB). The ACB is located in the concrete-lined charcoal bed cell which is below ground level located outside the MSRE building. Therefore, there was a concern for the potential of a nuclear criticality accident, although water would have to leak into the chamber for a criticality accident to occur. Unstable carbon/fluorine compounds were also formed when the fluorine reacted with the charcoal in the charcoal bed. The purpose of the proposed measurement system was to perform an accountability measurement to determine the fissile mass of ^{233}U in the primary vessel. The contents of the primary containment assembly will then be transferred to three smaller containers for long term storage. Calculations were performed using MCNP-DSP(4) to determine the configuration of the measurement system. The information obtained from the time signatures can then be compared to the measurement data to determine the amount of ^{233}U present in the primary containment assembly.

The Molten Salt Reactor Experiment Facility (MSRE) operated from 1965 to 1969. The fuel was a molten salt that flowed through the reactor core which consisted of uranium tetrafluoride with molten lithium and beryllium salt used as the coolant. In 1968 the fuel was switched from ^{235}U to ^{233}U . The Molten Salt Reactor Experiment was canceled in 1969 at which time approximately 4800 kg of salt was transferred to the fuel drain tanks. There was about 36.3 kg of uranium, 675 grams of plutonium and various fission products present in the fuel salt.⁽³⁾ The salt was allowed to solidify in the fuel drain tanks. The salt was heated on a yearly basis to recombine the fluorine gas with the uranium salt mixture. In 1990 this process was ceased to conduct an investigation to determine the possible problem of radioactive contamination. In March 1994, a gas sample was taken from the off gas system that indicated ^{233}U had migrated from the fuel drain tank system to the off gas system. It was found that approximately 2.6 kg of uranium had migrated to the Auxiliary Charcoal Bed (ACB). The ACB is located in the concrete-lined charcoal bed cell which is below ground level located outside the MSRE building. Therefore, there was a concern for the potential of a nuclear criticality accident, although water would have to leak into the chamber for a criticality accident to occur. Unstable carbon/fluorine compounds were also formed when the fluorine reacted with the charcoal in the charcoal bed. These components could cause an explosion at elevated temperatures.⁽¹⁾ A system was placed at MSRE to remove the reactive gases in 1995 and 1996. Once the reactive gases are removed the solid uranium deposits will be removed from the charcoal bed. The fabrication and installation of the Charcoal Bed Mockup that will be used to simulate the charcoal removal process is complete.⁽¹⁾

The ACB consist of four tubes made of six-inch schedule 10 stainless steel. The tubes are approximately twenty-four inches long and connected in series. The estimated volume of the four tubes is 506 liters with the deposit containing approximately 2.6 kg of uranium. The bulk density of the laden charcoal is 0.44 g/cm^3 . Once the charcoal deposit is removed a passivation process will be performed to remove any excess impurities in the primary containment assembly. The isotopic contents of the laden charcoal after passivation in shown in Table 1.

Table 1. Isotopic Composition of Charcoal after Passivation

Element	Weight (g)	Molecular Weight	Moles	Mole Percent
Carbon	2675	12.01	35.7	66.14
Ash ^a	71	28.1	3	0.76
Uranium	2600	233	11	3.31
Fluorine	1906	19.0	100	29.79
Nitrogen	780	14.01	56	9.05
Hydrogen	224	1.01	223	36.21
Total	8256		428.7	

^a It was assumed that the average atomic weight of Ash is similar to that of Silicon.

The bulk density of the laden charcoal after passivation is 1.32 g/cm^3 . Table 1 assumes there is 2.6 kg of ^{233}U in the ACB. This estimated amount of ^{233}U is not necessarily the exact amount of uranium contained in the charcoal bed. Therefore, the isotopic composition of the laden charcoal had to be calculated with varying masses of uranium present. Since the ^{233}U fuel is a uranium tetrafluoride-lithium eutectic that was added to the fuel carrier salt during loading of the fuel, then UF_4 has to be taken out of the

isotopic composition. For each mole of uranium present there is also four moles of fluorine associated with it; therefore the amount of fluorine also has to be adjusted. The amount of carbon, ash, nitrogen and hydrogen will remain constant in the system and will not be affected by the amount of ^{233}U present. The density of the laden charcoal changes as the amount of ^{233}U changes. The density was calculated using

$$\rho_{\text{mix}} = 1/\sum(wf_i/\rho_i) \quad (1)$$

The purpose of the proposed measurement system was to perform an accountability measurement to determine the fissile mass of ^{233}U in the primary vessel. The contents of the primary containment assembly will then be transferred to three smaller containers for long term storage.

The primary vessel is a criticality safe container made of schedule forty stainless steel with an outside diameter of 11.43 centimeters and a height of 141 centimeters. A measurement system using a ^{252}Cf source, ^3He detectors, lead and polyethylene were designed to determine the fissile mass removed from the charcoal bed. Californium-252 is a spontaneously fission source that emits neutrons with an average energy of 2.13 MeV that are thermalized using polyethylene since ^3He detectors detect thermal neutrons only. A ^{252}Cf source is located on one side of the primary containment assembly and thirteen 2.54-centimeter diameter ^3He detectors are located outside the aluminum support rings. Horseshoe shaped aluminum support rings surround the primary containment assembly for stabilization of the system. A sketch of the measurement system is shown in Figures 1 and 2. The gamma dose rate is estimated to be approximately 800R/hr in the auxiliary charcoal bed near the ^{233}U deposit. Therefore, lead is placed between the aluminum support rings to reduce the dose to the detectors because ^3He detectors can

only withstand about 200 R/hr. The dose at the detectors was calculated to be 5 R/hr using MCNP4a.(2)TM

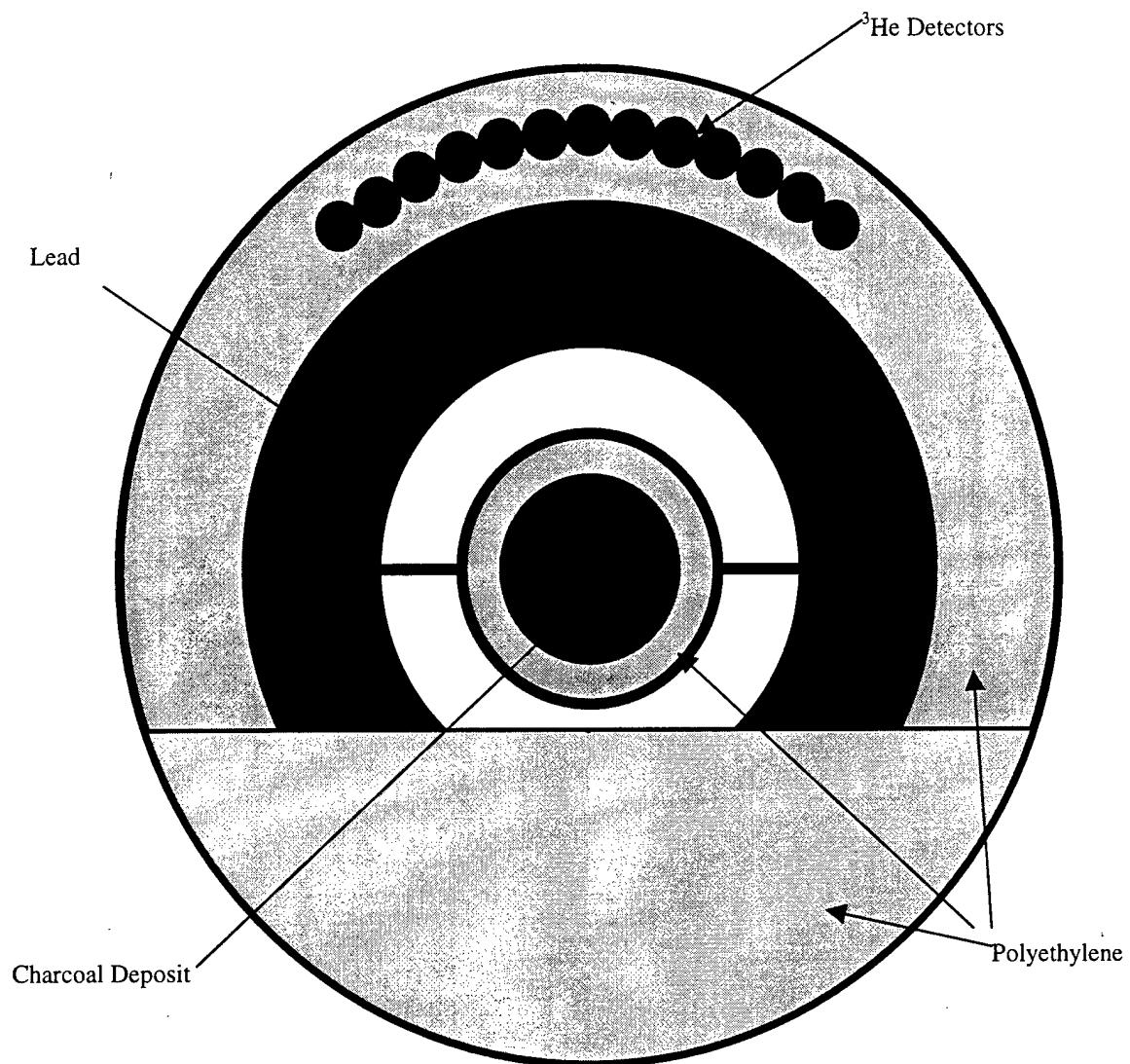


Figure 1. Horizontal View of Measurement System

Calculations were performed using MCNP-DSP(4) to determine the configuration of the measurement system. MCNP-DSP is a modified version of MCNP that is used to simulate source-driven correlation measurements that has several features that are

beneficial in designing this type of measurement system. The code uses a dual particle source which produces time-correlated neutrons and gamma rays. Variance reduction is

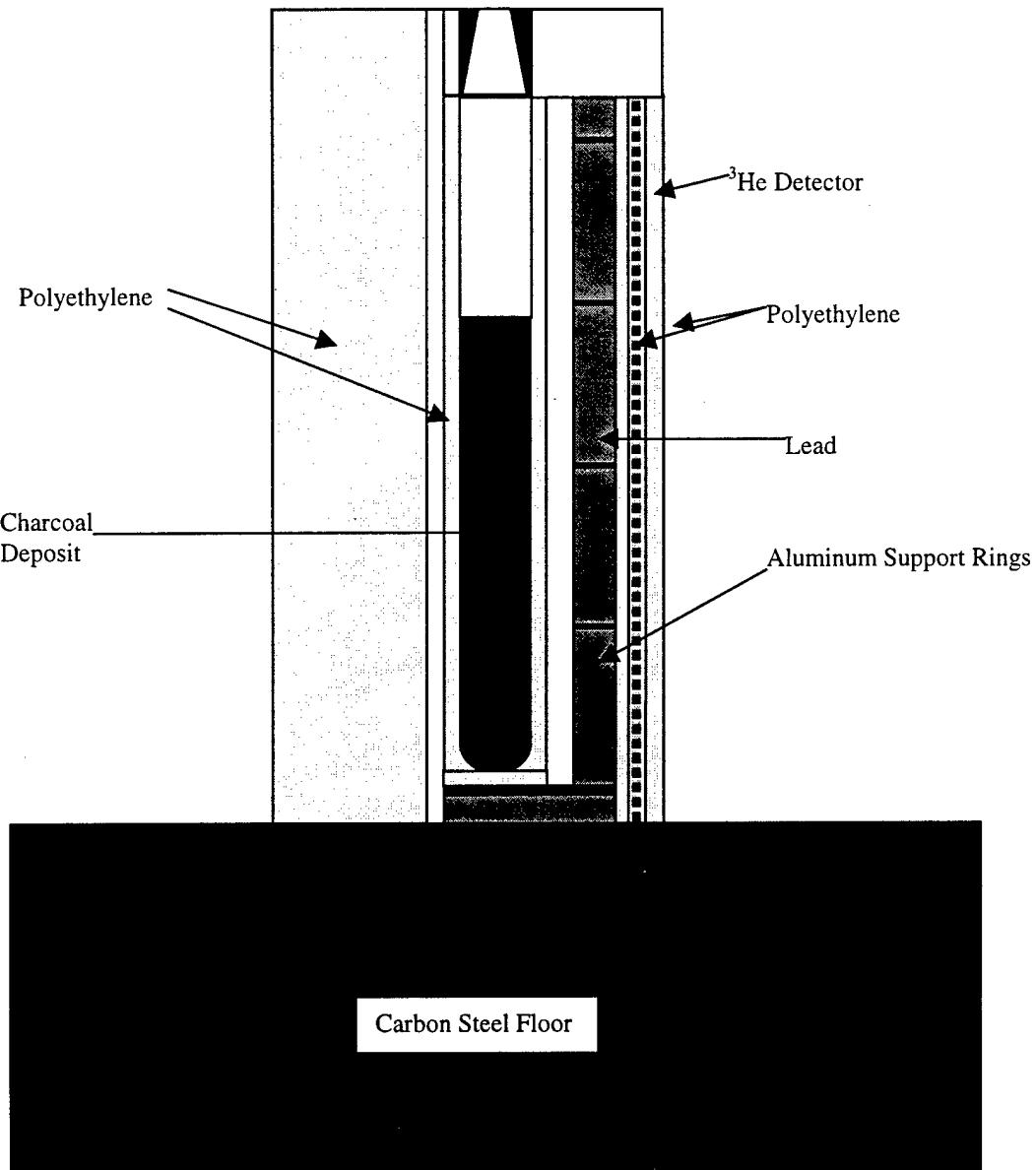


Figure 2. Vertical View of Measurement System

not used in MCNP-DSP to make use of analog particle tracking to represent the collision physics on an event-by-event basis. When the average quantities are used, such as \bar{v} , the fission chain fluctuations are reduced. Therefore, probability distributions replace average parameters because the spectral densities provide statistical estimates of the fission chain fluctuations. The proper treatments of angular distribution of neutrons from fission are included in the code to adequately describe the particle direction from both spontaneous and induced fission. MCNP-DSP uses detector specification in the input file. (5) The ^3He detectors specified in the input file are capture detectors in which the detector response is from the absorption of a neutron and the emission of secondary charged particles. The charged particles ionize the ^3He and produce an electrical pulse that is proportional to the kinetic energy of the secondary charged particle. For every neutron absorbed a count is scored in the appropriate time bin. Each time bin in the calculation corresponds to one nanosecond of real time.

Because ^3He detectors are thermal neutron detectors, polyethylene was suggested to thermalize the neutrons before detection. MCNP-DSP calculations were used to determine the amount of polyethylene for the measurement. The polyethylene thickness around the detectors was varied from zero to 3.81 centimeters. The number of neutrons detected versus polyethylene thickness is presented in Figure 3. A polyethylene thickness of 2.54 centimeters yielded the maximum number of neutrons detected by the 2.54-cm diameter ^3He detectors.

Once the amount of polyethylene surrounding the detectors was optimized, several calculations were performed with 2.54 centimeters of polyethylene surrounding

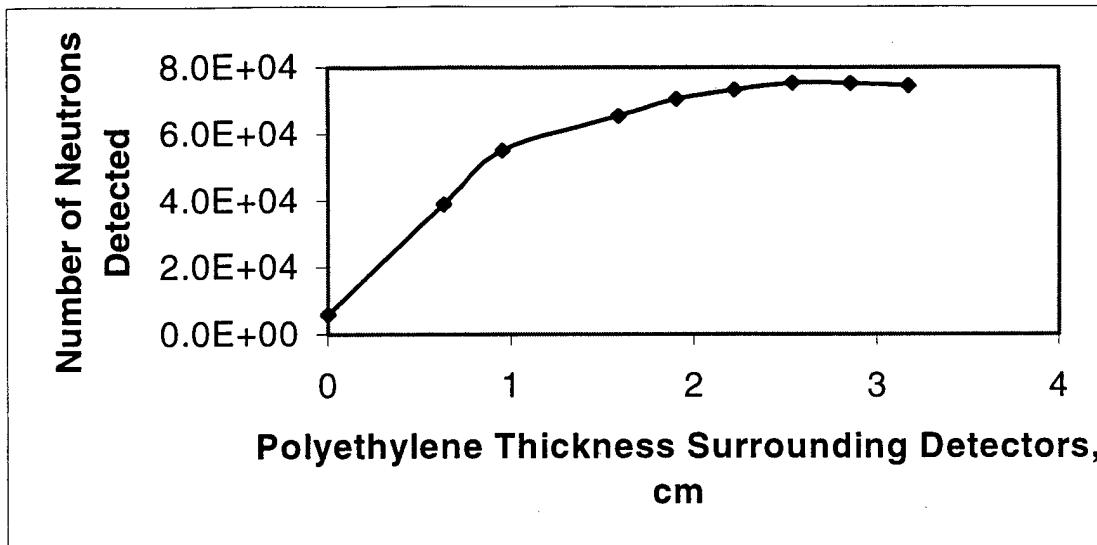


Figure 3. Number of Neutrons Detected as a Function of Polyethylene Thickness Surrounding Detectors

the detectors present and the amount of polyethylene surrounding the ^{252}Cf source varying from zero to 3.81 centimeters. The maximum number of neutrons detected occurred when no polyethylene surrounded the ^{252}Cf source. The results of these calculations are presented in Figure 4.

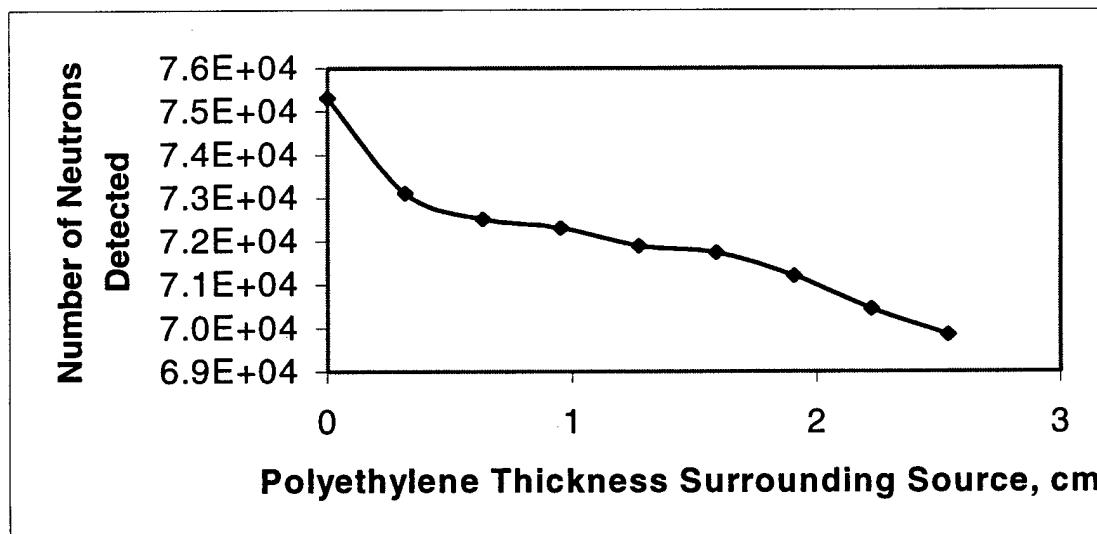


Figure 4. Number of Neutrons Detected as a Function of Polyethylene Thickness Surrounding Source

Several time domain signatures were calculated with various masses of ^{233}U present in the primary containment assembly. The randomly pulsed neutron measurement with ^{252}Cf is a measurement of the impulse response in the system. This type of measurement is simulated by starting the ^{252}Cf source particles at time zero and tracking the source particles and their progeny to the detectors.(5) The detection time for each particle is determined and counted for the appropriate time bin. The time bins are then incremented similar to the pulse height spectra with a MCA. There was not much sensitivity in the signatures obtained from MCNP-DSP. Therefore, calculations were performed using MCNP4a to determine k_{eff} of the system as a function of mass of ^{233}U . k_{eff} varied in the system between 0.11 and 0.26. The results are shown in Figure 5.

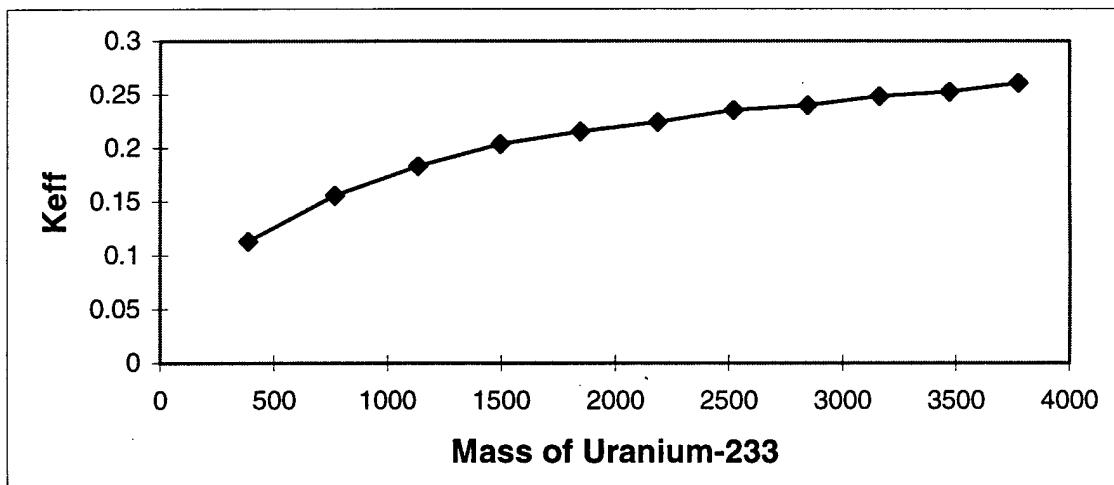


Figure 5. k_{eff} of System as a Function of Uranium Mass

To increase the k_{eff} of the system polyethylene was added around the primary containment assembly to serve as a reflector. It is desired for the polyethylene not to extend past the front side of the primary containment assembly. The maximum amount of polyethylene possible is 3.175 cm. This amount of yielded a k_{eff} of 0.527 ± 0.00235 .

The k_{eff} results as a function of polyethylene thickness surrounding the primary containment assembly is shown in Figure 6.

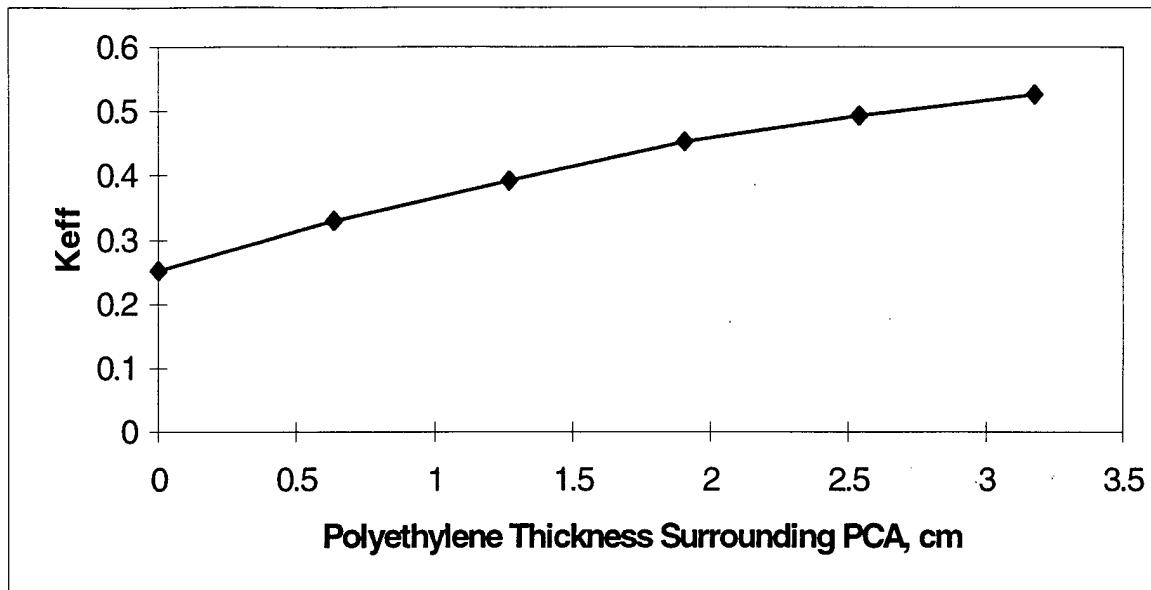


Figure 6. K_{eff} of System as a Function of Polyethylene Thickness Surrounding PCA

Approximately 58 percent of the neutrons were escaping the measurement system. Therefore, the front of the measurement system was enclosed with polyethylene to reflect neutrons back into the system. With the front of the system enclosed with polyethylene the percent of neutrons leaking from the system is reduced to 6 percent. The percent of fission neutrons also increased from 16 to 20 percent.

Times domain calculations were completed using the final measurement system design. The mass of ^{233}U present in the system ranges from 767 to 3472 grams. The results of the time signatures are shown in Figure 7.

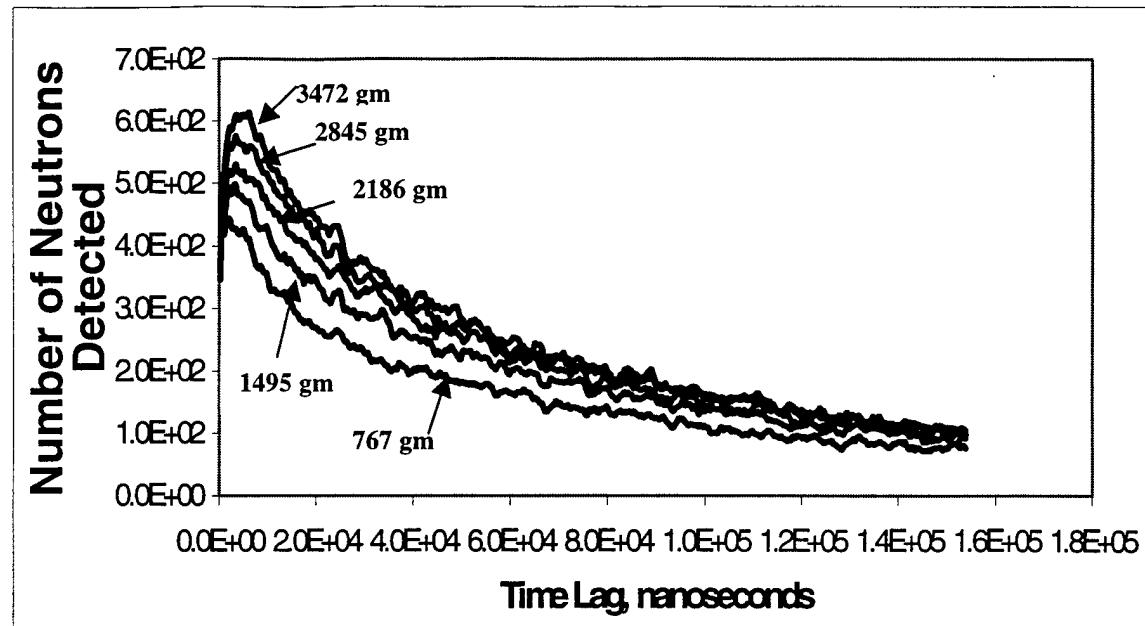


Figure 7. Time of Flight Signatures

The signatures for the varying amounts of ^{233}U in the system can be seen clearly during the first few seconds of measurement time. The integral values of the signatures are shown in Table 2.

Table 2. Integral Value of Time of Flight Signatures

$^{233}\text{U, grams}$	Integral Value
767	6.4E+04
1495	7.8E+04
2186	8.7E+04
2845	9.3E+04
3472	9.7E+04

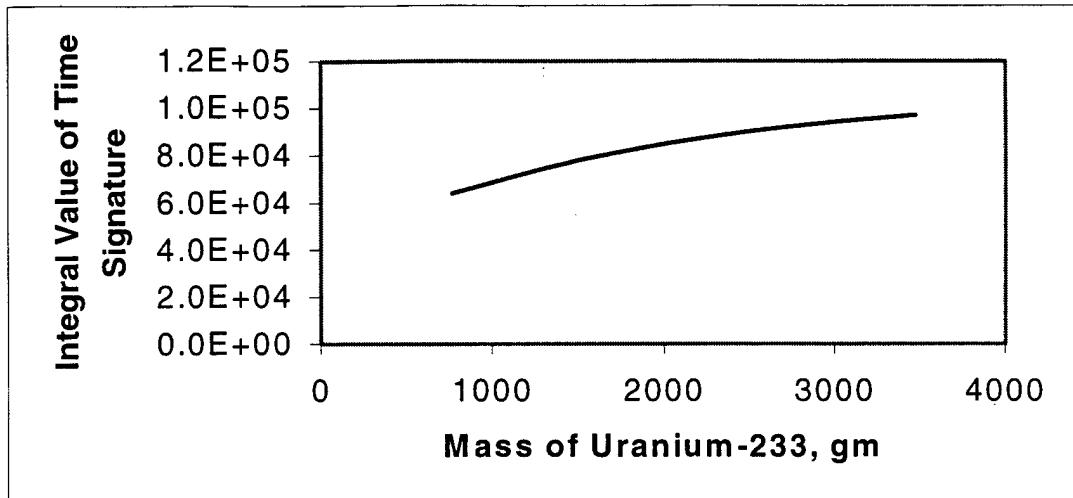


Figure 8. Integral Value of Time Signature as a Function of Uranium Mass

It is shown that there is a noticeable difference between the integral values of the time signatures as the mass of ^{233}U in the system changes. Therefore, this measurement system design should be efficient in determining the amount of ^{233}U removed from the ACB.

To calibrate the measurement system, measurements on an experimental mockup containing laden charcoal with ^{235}U will be performed. Two separate calculations will be performed using MCNP-DSP on the experimental mockup containing laden charcoal with ^{233}U and laden charcoal with ^{235}U . The ratios of the calculations will then be compared and the measured value of the calibration factor with ^{233}U can then be obtained.

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