FINAL SCIENTIFIC/TECHNICAL REPORT FOR SBIR PHASE-I

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Recipient: Reeves and Sons LLC

Title: **A Device to Measure Low Levels of Radioactive Contaminants in Ultra Clean Materials.**

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SUMMARY

The purpose of this research was to develop a radiation detection device so sensitive that a decay rate of only one atom per 11.57 days per kilogram of material could be detected. Such a detector is needed for screening materials that will be used in exotic high-energy physics experiments currently being planned for the near future.

The research was performed deep underground at the Underground Mine State Park in Soudan, Minnesota. The overburden there is ~1800 meters water equivalent (MWE). The reason for performing the research at such depth was to vastly reduce the effects of cosmic radiation. The flux of muons and fast neutrons is about 100,000 times lower than at the surface. A small clean room quality lab building was constructed so that work could be performed in such a manner that radioactive contamination could be kept at a minimum. Glove boxes filled with dry nitrogen gas were used to further reduce contamination from dirt and also help reduce the concentration of the radioactive gas ^{222}Ra and daughter radionuclides, which are normally present in air.

A massive lead shield (about 20 tons) was constructed in such a manner that an 8 inch cube of space in the center was available for the sample and detector. The innermost 4 inch thick lead walls were made of ~460 year old lead previously used in double beta decay experiments and known to be virtually free of ^{210}Pb. A 1.5 inch thick shell of active plastic scintillator was imbedded in the center of the 16 inch thick lead walls, ceiling, and floor of the shield and is used to help reduce activity due to the few muons and fast neutrons seen at this depth. The thick lead shielding was necessary to shield the detector from gamma rays emitted by radionuclides in the rock walls of the mine. A sealable chamber was constructed and located on top of the shield that included a device for raising and lowering the detector and samples into and out of the center chamber of the shield.

A plastic scintillator detector measuring 6x6x6 inches was fitted with wavelength shifting fibers that allowed the light from ionizing radiation to be collected and transmitted outside the massive shield to photomultiplier tubes and electronics. The detector was calibrated for energy and detection efficiency and low-resolution background spectra were collected.

Results from these measurements show the figure of merit (using: efficiency/square root of background) for this plastic scintillation counting technique to be ~15 times better than for a 2 kg germanium detector for measuring surface contamination from atmospheric ^{222}Rn daughters (^{210}Pb, ^{210}Bi, and ^{210}Po). These daughter radionuclides are normally deposited **everywhere** onto all materials exposed to air.

The results are encouraging and indicate that plastic scintillation counting techniques can be of benefit to the public by making available very sensitive counters for screening ultra-low background materials at an affordable cost. However, in order to reach the level required a multi element array of thin plastic scintillator sheets must be developed that will allow many thin samples to be counted at one time. In addition, more sophisticated light detection hardware, electronics, and computer software is needed.

3 Executive Summary: 1) No Significant additions to the understanding of ultra-low background counting have as yet been discovered. **2)** The effectiveness and feasibility of the method are positive and encouraging. The figure of merit (using: efficiency/square root of background) for this plastic scintillation counting technique is currently ~15 times better than for a 2 kg germanium detector for measuring surface contamination from atmospheric ^{222}Rn daughters (^{210}Pb, ^{210}Bi, and ^{210}Po). These daughter radionuclides are normally deposited **everywhere** onto all materials exposed to air. **3)** No benefit to the public has yet been demonstrated but the tentative results are encouraging and indicate that plastic scintillation counting techniques can be of benefit to the public by making available very sensitive counters for screening ultra-low background materials at an affordable cost.

4 Comparison of Goals to Accomplishments: The objective was to build a counting system that would measure 1 micro Becquerel per kilogram of material. The goals were as follows: **1)** Gain access to an underground site. **2)** Construct a clean underground lab building (clean room quality). **3)** Build a massive ultra-low background lead shield containing at least one layer of active plastic scintillator to be used for detecting cosmic ray induced events. **4)** Build a lifting/lowering device located on top of the shield and contained inside a radon resistant chamber (equipped with glove ports, gloves, and docking port) that allows samples to be introduced to the chamber without being contaminated with radon and then be stored or loaded and lowered into the counting chamber located at the center of the massive lead shield. **5)** Build a number of nitrogen glove boxes (with docking ports) for handling samples. **6)** Design and build transport vessels that dock to each docking port located on glove boxes, detector shield, or electroforming modules. **7)** Develop a vacuum system to allow the transport chambers and docking ports to be evacuated and backfilled with cover gas when necessary to reduce radon contamination. **8)** Design and build a 50-element plastic scintillator detector that would hold 51, 1 mm thick samples of copper, lead, plastic, electronic parts, or whatever. Design and implement the necessary electronics to handle the signals from the main detectors and the anticoincidence signals from the plastic scintillator shell located midway between the detector and the outside of the lead shield. **9)** Test and evaluate the detector. **10)** Write and submit the final report.

Goals 1 through 7 were met although a no cost, 6 month extension was needed. Figure 1 shows the outside of the underground lab building located at ~1800 Meters Water Equivalent (MWE) in the Soudan Underground Mine State Park at Soudan, MN. A 160 liter liquid nitrogen Dewar that supplies the nitrogen cover gas that helps mitigate the radon problem can be seen. A nitrogen hood (docking port not yet installed) is shown in Figure 2.

Figure 1 - Lab Building at Soudan Underground

Figure 2 - Nitrogen Glove Box

Figure 3 shows the lead shield wrapped with aluminum foil to help keep radon out with the lifting/lowering device installed (the front, which contains glove ports is not in place and the docking port has not yet been installed).

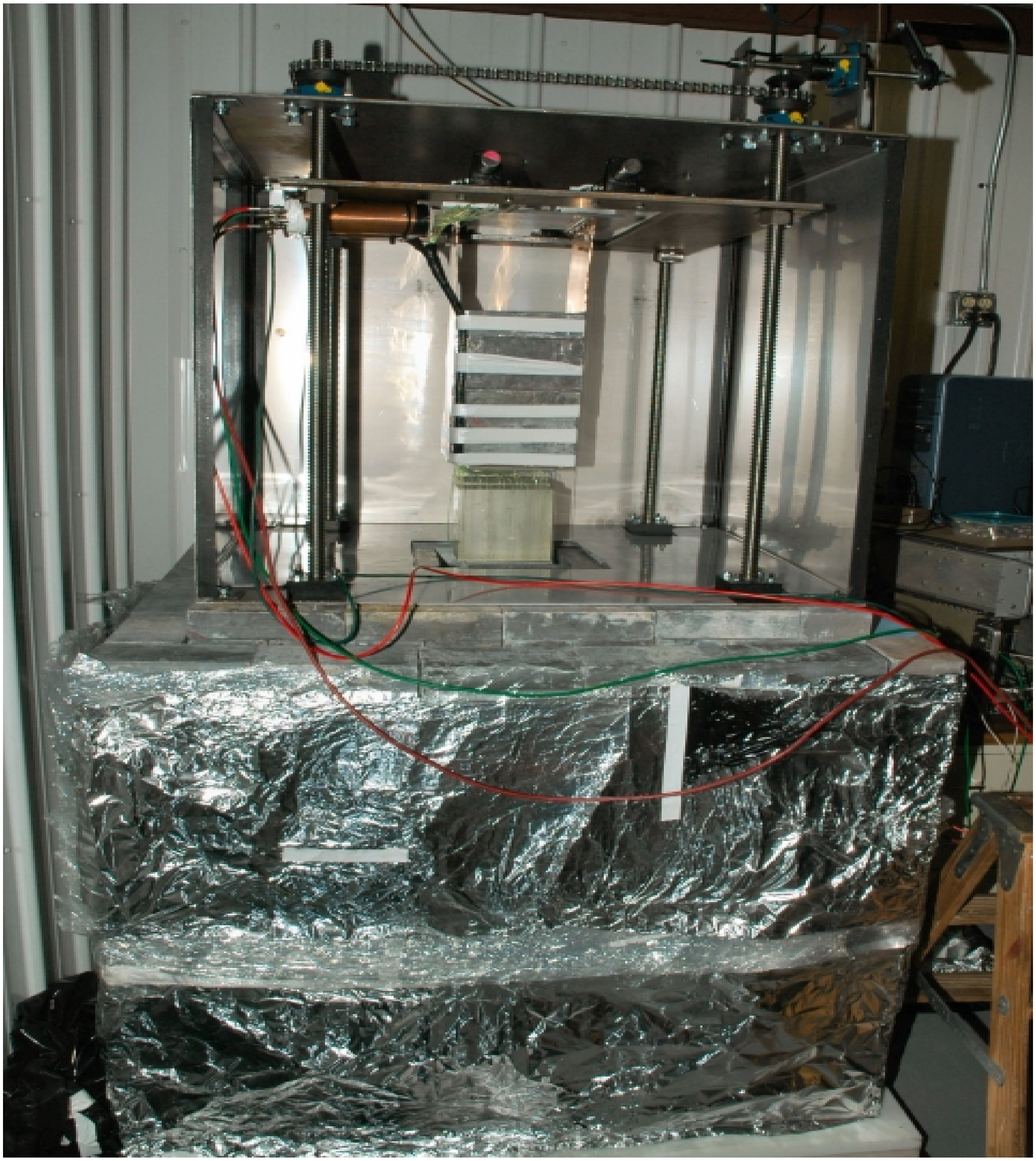

Figure 3 - Lead shield with lifting/lowering device and plastic scintillation detector, ready to be lowered into the shield cavity.

A Varian vacuum system containing a dry scroll roughing pump and turbo supplies the vacuum needed to expel radon containing air from the transport vessels and docking ports and allow efficient introduction of a nitrogen cover gas.

Figure 4 shows the 15 cm cube of plastic scintillator, the small diameter wavelength shifting fibers, and photomultiplier tubes fitted with copper tubes to shield EM interferance.

Figure 4 - Plastic scintillation detector (6"x 6"x6") with wavelength shifting fibers and photomultiplier tubes (housed in copper shielding tubes at the upper left)

Goal 8 (one of the most important goals) was not met. The 50 elements for the plastic scintillator detector are on hand but have not yet been assembled and tested. In the interim, a 15 cm cube of plastic Scintillator, outfitted with 36 wavelength shifting fibers, and photomultiplier tubes was installed inside the lead shield and tested (the innermost 4 inch thick layer of lead is comprised of the ~ 460 year old lead used during the early stages of IGEX at Homestake Mine in Lead, SD and known to have been virtually free of ^{210}Pb. The background obtained is shown in Figure 5, which also shows the background attained with germanium detectors during the IGEX double beta decay experiments. The ultimate efficiency of this device for measuring ^{210}Pb (through the beta emitting daughter ^{210}Bi) compared to the efficiency of a 2 kg germanium detector is depicted by the Monte

Carlo spectra shown in Figure 6. The overall efficiency for detecting surface contamination of ^{210}Pb (through its daughter radionuclides ^{210}Bi and ^{210}Po) with a 15 cm cube of plastic scintillator should approach 100 percent (~ 50% for the ^{210}Bi beta and ~50% for the ^{210}Po alpha – both of which are generally present) which would be ~500 times higher than the efficiency of a typical 2 kg germanium detector for detecting these radionuclides (the efficiencies are based on the total counts in the Monte Carlo simulations from 600,000 ^{210}Bi decays with the counts seen by the plastic scintillator being doubled since virtually all alphas striking it would also be detected. The production of bremsstrahlung from the ~1200 keV ^{210}Bi beta is very low and typical germanium detectors detect NEITHER alphas NOR betas due to the rather thick materials used to construct the vacuum cryostats necessary to house the germanium diode detectors. Sensitivity is a function of both detector counting efficiency and background so the relative merits of the detectors must include both. A germanium detector measures **ONLY** the bremsstrahlung produced by ^{210}Bi from the decay of ^{210}Pb while the plastic scintillator detector measures the bremsstrahlung in addition to measuring betas from ^{210}Bi and alphas from ^{210}Po**.**

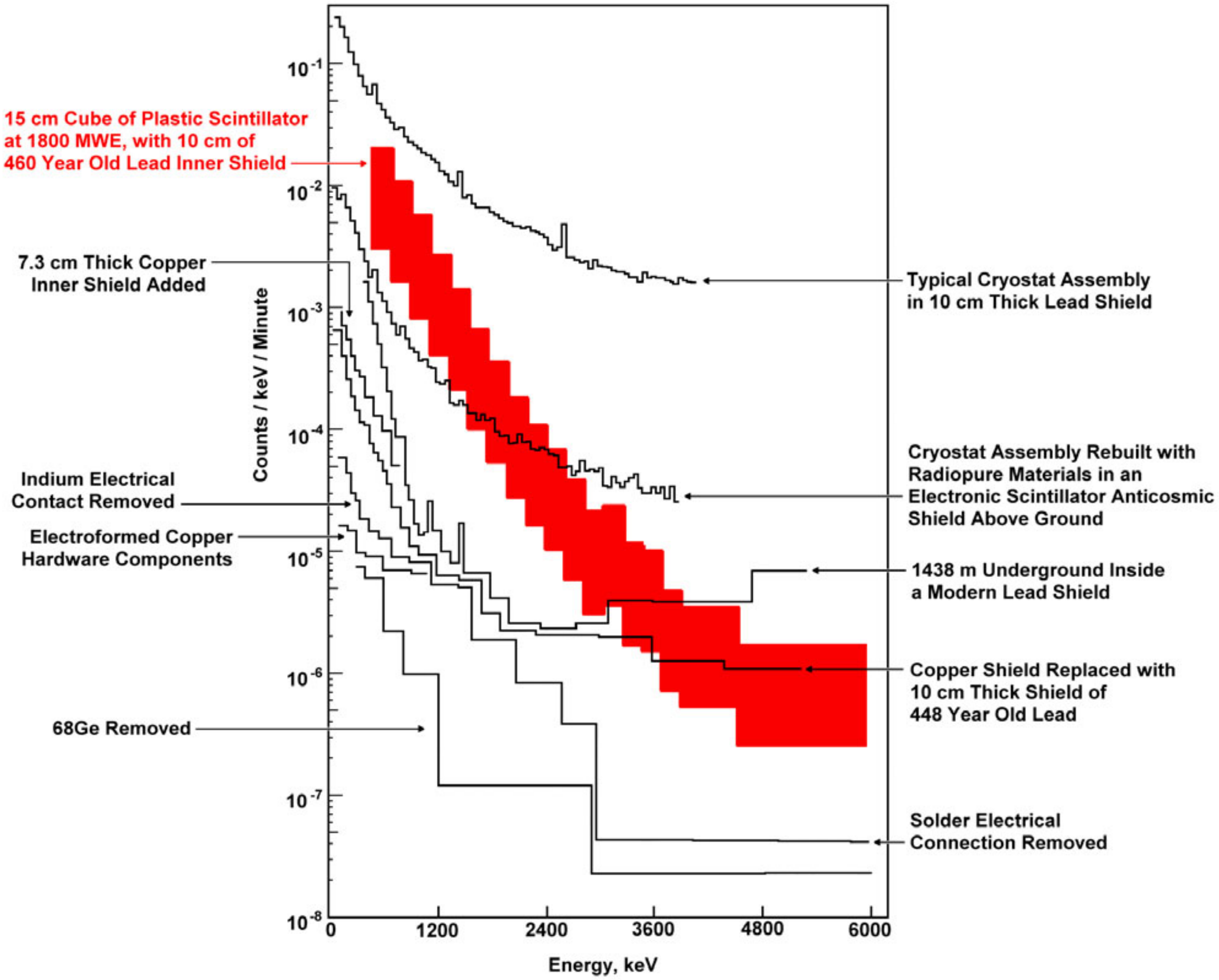

Figure 5 - Background of a 15 cm cube plastic scintillation detector (broad red line due to uncertainties in energy and efficiency calibrations) taken at The Soudan Underground Mine (~1800 MWE) compared to background spectra taken during a decade of development of the germanium detectors used in the IGEX double beta decay experiment in the Homestake mine.

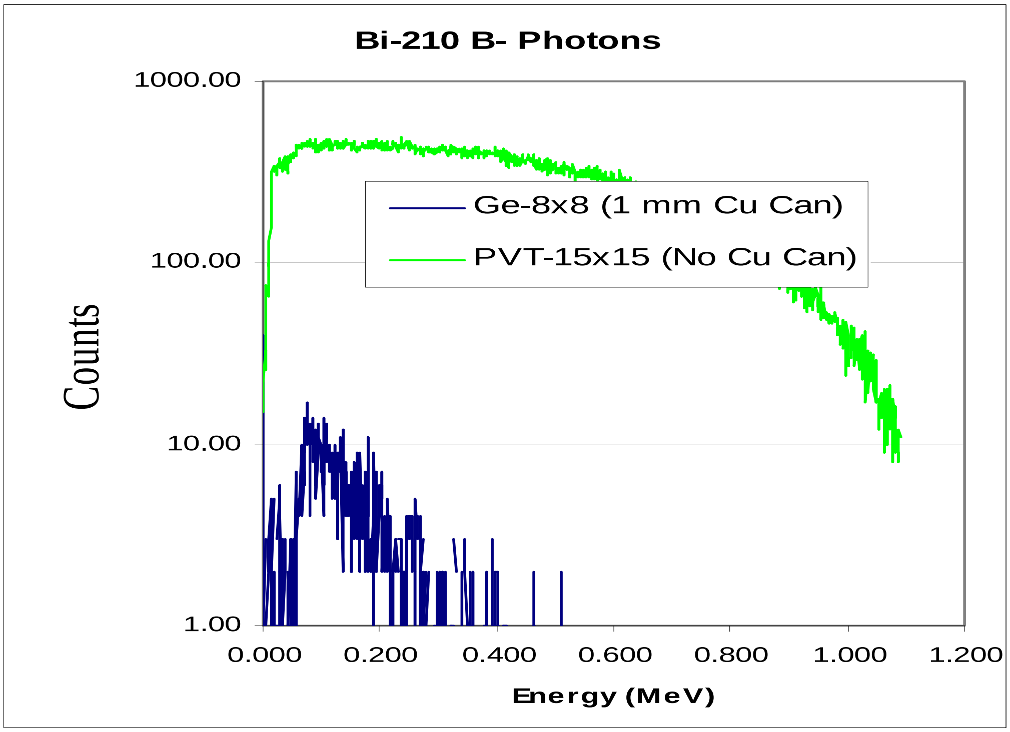

Figure 6 - MCNPX Monte Carlo generated spectra from 600,000 decays of ^{210}Bi for a 15 cm diameter by 15 cm long plastic scintillation detector and a 2 kg germanium detector.

Limited energy and efficiency calibrations have been performed, but due to the extremely low resolution of the detector, the uncertainty is large and more work needs to be done for both energy and efficiency calibrations to reduce the uncertainties. **The actual efficiency that we obtained for gamma rays (as measured with ^{40}K) is ~ ¼ that predicted by Monte Carlo simulations for ^{40}K (due to light losses from using the wave shifting fibers and to dead time from high count rates resulting from photomultiplier tube noise that must be operated in coincidence in order to separate the signal of interest from phototube noise)**. These light losses reduced the signal from the ionizing radiation relative to the noise arising from the phototubes and forced a higher energy threshold to be used that excluded about half the counts due to the ^{40}K. However, we believe the counting efficiency for alphas and betas is very high for surface contamination since the energy deposited should be sufficiently high to be seen above the noise even with the light losses incurred from using wave length shifting fibers. We have a very limited supply of radioactive sources at the underground site and have not yet been able to calibrate the detector as well as we would like. Since counting efficiency is a linear factor while background is a square root function, the improvement of a factor of ~500 in counting efficiency over that of a 2 kg germanium detector more than compensates for the increased background which is currently ~100 times higher than that of a superbly constructed 2 kilogram germanium detector. Work continues on understanding the difficulties and significance of counting alphas, betas, and gammas at high efficiency and to find ways to further reduce the background and improve the counting efficiency. When implemented, the 50-element array of thin plastic scintillation sheets surrounded by sample should further reduce the system background while maintaining or improving the counting efficiency. Background coming from contamination of the inner surfaces of the lead shielding will be reduced by the outermost sample sheets (the innermost lead used was at one time free of surface contamination but spent ~10 years underground protected only by thin plastic (it is expected that ^{210}Pb, ^{210}Bi, and ^{210}Po from radon are currently on the surfaces). If the background for each individual plastic scintillating sheet is sufficiently low, certain radionuclides from Th and U decay chains can be identified through timing coincidence (some radionuclides in the chains have half-lives of less than 1 sec, which means they will decay shortly after the parent decays).

5 Summary of project activities: The original hypotheses was that (due to low counting efficiency caused by self absorption and solid angle restraints) samples of high density and high atomic number could not be readily measured at the 1 micro Bq. per kg level with a single (even though large) low background germanium detector and that the cost of a large array of low background germanium detectors would be very high. Using many thin samples sandwiched between thin (low cost) plastic scintillation elements would allow a very sensitive detector to be constructed at an affordable cost. It was known that two major problems would have to be overcome (radioactive contamination and low light yield).

The proposed approach was to mitigate radioactivity produced by photomultiplier tubes by using wave length shifting fibers to collect light and then carry it to the outside of a massive lead shield through small diameter light pipes. Cosmic ray interaction would be virtually eliminated by locating the detector deep underground. The signals from plastic scintillation panels embedded in the thick lead shield can be used, via coincidence, to reduce the activity caused by the few muons passing through the shield at this depth and reduce the activity caused from muon induced fast neutrons produced in the rock walls and ceiling. Performing final assembly and modifications inside nitrogen glove boxes and not exposing either detector or samples to air would reduce surface contamination. If the amount of radioactivity inherent to the scintillation panels themselves was found to be significant, scintillation panels could possibly be produced on site from clean starting materials.

Permission was obtained from The University of Minnesota, The State of Minnesota Department of Natural Resources, and Fermi Lab (all of whom had to agree) to allow us to locate the necessary materials and equipment underground at The Soudan Underground Mine State Park at ~1800 MWE located at Soudan, MN. All three parties were very cooperative and helpful. Despite their cooperation, it still took many months and many man-hours to complete all the paperwork, planning, meet all the safety concerns, and get all the necessary signatures.

Lead for the massive shield and old plastic scintillation modules that could be used for active coincidence were being stored at PNNL, awaiting excess. Responsible scientists at PNNL had been contacted prior to submitting the Phase-I proposal and had agreed this would be a good use for both and were eager to proceed with the transfer. However, getting the transfer of the lead from PNNL to Soudan to actually take place took so long that a 6 month extension to the Phase-I project was necessary. Almost one ton of ~460 year old lead was “borrowed” from the old IGEX experiment on a temporary basis and was used for the inner four inches of shielding. It became obvious after several months that it was unlikely we would obtain the plastic scintillation panels being stored at PNNL in time to do the machining and use them in the shield so new panels were purchased (the PNNL scintillation modules continue in outdoor storage but we have not been able to get them transferred).

The detector lifting device, nitrogen glove boxes, and other items were fabricated in Richland, WA and transported to Soudan. A small building was constructed on site at Soudan. A vacuum system was delivered directly to Soudan. The massive lead shield was constructed with a 1.5 inch thick layer of plastic scintillation modules imbedded between the inner 8 inches of lead and the outer 8 inches of lead. The scintillation panels had been fitted with wave shifting fibers and were connected to photomultiplier tubes outside the lead shield. The lifting device was installed along with a detector consisting of a 6 inch cube of plastic scintillation material fitted with 36 wavelength shifting fibers and connected to photomultiplier tubes (outside the lead shield). Some testing and evaluation has been performed.

There were no major departures from the planned methodology other than timing. The long time and the many hours required to obtain site permission and arrange for the lead, definitely had a detrimental impact on the project. It had been expected that a significant length of time would be required but the actual time was far more than had been anticipated (especially for getting the lead from PNNL). The time and effort required to accomplish the various technical tasks, designing and building the many components for the various pieces of equipment were also far more than had been anticipated. To have fully accomplished all that had been planned would have required Phase-I funding and a significant fraction of a fully funded Phase-II. Although at a slow pace, work continues whenever practical at the underground site to assemble, calibrate, and test, the 50-element array of plastic scintillation sheets.