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# Development of Neutron Probes for Characterization of Hazardous Materials in the Sub-surface Medium

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## ABSTRACT

Neutron probes are being developed at the Idaho National Engineering and Environmental Laboratory (INEEL) for the detection, identification and quantification of hazardous materials in the ground. Such materials include plutonium, uranium, americium, chlorine and fluorine. Both a Neutron Gamma (NG) probe and a Prompt Fission Neutron (PFN) probe are being developed. The NG probe is used primarily for nuclide identification and quantification measurements. The PFN is used mostly for the detection and measurement of fissile material, but also for the determination of thermal neutron macroscopic absorption cross sections of the various elements comprising the ground matrix. Calibration of these probes will be carried out at the INEEL using an indoor facility that has been designed for this activity.

## I. INTRODUCTION

The Nuclear and Radiological Sciences group at the Idaho National Engineering and Environmental Laboratory is developing neutron generator probes for elemental analysis of the subsurface at the INEEL. The probes are part of a research and development effort to locate, identify and quantify elements such as plutonium, uranium, americium, carbon, chlorine and fluorine in the ground. These efforts support the characterization and remediation of buried waste sites at the INEEL. Each probe consists of a neutron generator and gamma/neutron detection system, as depicted in figure 1. Presently, the two probes being developed are a Neutron Gamma (NG) probe and a Prompt Fission Neutron (PFN) probe. The NG probe is used primarily for elemental analysis and quantification measurements, while the PFN probe is used for the detection and measurement of fissile materials, including the determination of the thermal neutron-macroscopic absorption cross section of the subsurface.

They are designed so that they can be operated in existing INEEL wells. The diameters of the NG probe and the PFN probe are 10cm (4.0in) and 11cm (4.3in) respectively. The length of the NG probe is 4.3m (14ft), while the PFN probe is 4.6m (15ft).

The NG and PFN probes use customized MF Physics models A-320 and A-211 14MeV neutron generators respectively. The expected neutron yield is about  $10^8$  neutrons/second in each case. The NG probe has a High Purity Germanium (HPGe) detection system, while the PFN probe uses either NaI or Helium-3 detectors. These probes are sealed in a pressure housing to allow their operation while immersed in water, and therefore can be used in wells where water is present.

The wells at the INEEL are categorized as either shallow probe holes as is the case at the shallow burial grounds, or vadose zone and groundwater monitoring wells that traverse the Snake River Plain aquifer. The shallow probe holes exist for the purpose of using geophysical logging techniques to obtain subsurface elemental analysis data in areas where suspect transuranic waste has been buried. These wells are usually between 4.6m (15ft) and 7.6m (25ft) deep, and have an internal diameter of about 11cm (4.5in). The vadose zone and groundwater monitoring wells are constructed so that surveys can be carried out to check for the movement of potential underground contamination. These wells are up

to 244m (800ft) deep, and have diameters that range from 10cm (4.0 in) to about 91cm (36in).

The probes can be operated in either passive or active mode. In the passive mode the probes operate with the generator shut off and the ambient radiation is measured. The active mode involves using the neutron generator to activate the surrounding media and make gamma measurements with the NG probe, or inducing fission in fissile materials with the PFN probe, and using a differential die-away technique.

## II. CALIBRATION

Much of the calibration will be carried out at the INEEL, using an indoor facility that has been built to simulate field conditions. This facility, located at the Test Reactor Area, has been converted from a decontaminated reactor pool by placing two vertical tubes on the bottom of the pool, and filling the surrounding space with pea-sized gravel. This gravel filled pit houses a 74cm (29in) tube, and a 152cm (60in) tube that are 5.2m (17ft) deep.

Calibration work is scheduled in FY-02 using the 74cm (29in) diameter tube with a set of doughnut-shaped Stackable Light Surrogates (SLSs) containing representative subsurface mediums. The SLSs are filled with media such as soil, sand or concrete to simulate the underground medium. Each one, see figure 2, has a

center annulus for the placement of a probe, using actual well casing. The distribution of elements in the ground can be simulated, by placing samples into one of four ports in each SLS. Up to five containers are then stacked on top of each other inside the 74cm (29in) tube, such that the center annulus in each surrogate container is aligned with each other, see figure 3, to simulate a well. Plans are underway to prepare by year end a tank assembly in the 60in hole for performing calibrations and other testing in water. This is an efficiency calibration that involves inserting the probe into the simulated well hole and taking data to determine the relationship between the sample distance, mass and the detector response.

### III. ANALYSIS TECHNIQUE

The NG probe will operate in both passive and active modes. Subsurface radioactive contaminants can be measured using the HPGe detector in passive mode. Radionuclides are identified using the energy and intensity of the gamma ray peaks from acquired spectra. Standard spectral fitting techniques will be used to find the peak areas for the nuclides of interest. A combination of the measured response function of the detector and a Monte Carlo gamma-ray transport model will be used to determine homogeneous elemental concentrations.

To measure non-radioactive subsurface contaminants i.e. elements, the probe will be used in “active” mode. The neutron generator will produce neutrons that will either be scattered or captured by the elements of interest. By acquiring different spectra over different time periods following the neutron pulse, the scattering  $[(n,n')]$  reactions], capture  $[(n,\gamma)]$  reactions] and decay processes can be separated. Once again, standard spectral fitting techniques will be used to find the peaks, peak centroids and areas of the acquired spectrum. For the active-mode measurements, however, a neutron transport calculation will have to be combined with the response function of the detector and a gamma-ray transport calculation to calculate the homogeneous elemental concentrations.

The PFN probe will also have both passive and active modes. The passive mode will be used to provide neutron background levels, but is not expected to be particularly informative. The active mode should be the more useful of the two. The fissile material measurements will be made using a standard differential die-away technique. In short, as fissile materials interact with thermal neutrons, they fission, providing neutron multiplication. These fission neutrons enhance the flux of epithermal neutrons following the neutron generator pulse. Tracking the decay time of the epithermal neutron component and combining this with other information about the probe environment (porosity, neutron poisons, etc), as well as

with neutron transport calculations, will allow for a measurement of fissile materials in the environment surrounding the probe. Additionally, the PFN probe will be usable as a moisture probe. While not ideally designed for this type of measurement, the probe should be reasonably sensitive to moisture. Finally, use of a suite of NaI detectors will allow for macroscopic cross section measurements.

For both the NG and PFN probes, calibration will be vital to the final results. Surrogates to simulate various geological formations will be used to validate the various transport calculations, and validate the analysis technique. The facility to be utilized provides ample flexibility to use various surrogates and perform tests on homogeneous and heterogeneous matrix formations.

#### **IV. CONCLUSIONS**

The authors anticipate that initial probe characterization will be completed in FY-2002, and that the detection sensitivity for several nuclides of interest will be known at that time. The calibration facility at the Test Reactor Area will be suitable for calibrating these probes in both homogeneous and heterogeneous formations. The probes are expected to be ready for

demonstration in the field at the INEEL by the end of September 2002. Calibration and field data will be forthcoming.

#### **ACKNOWLEDGEMENTS**

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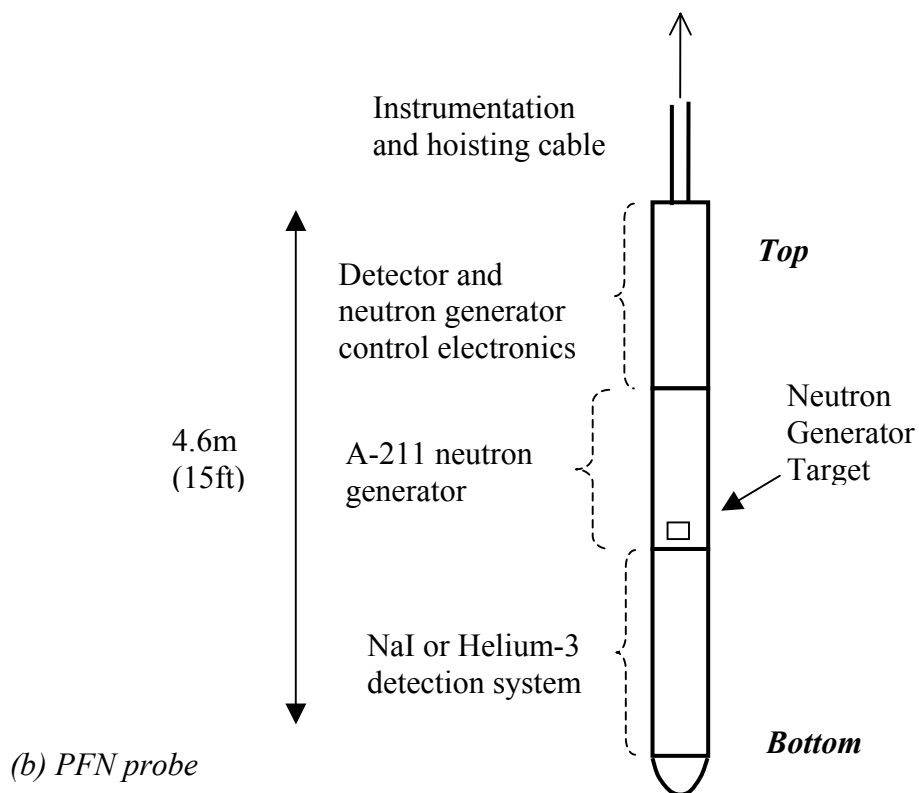
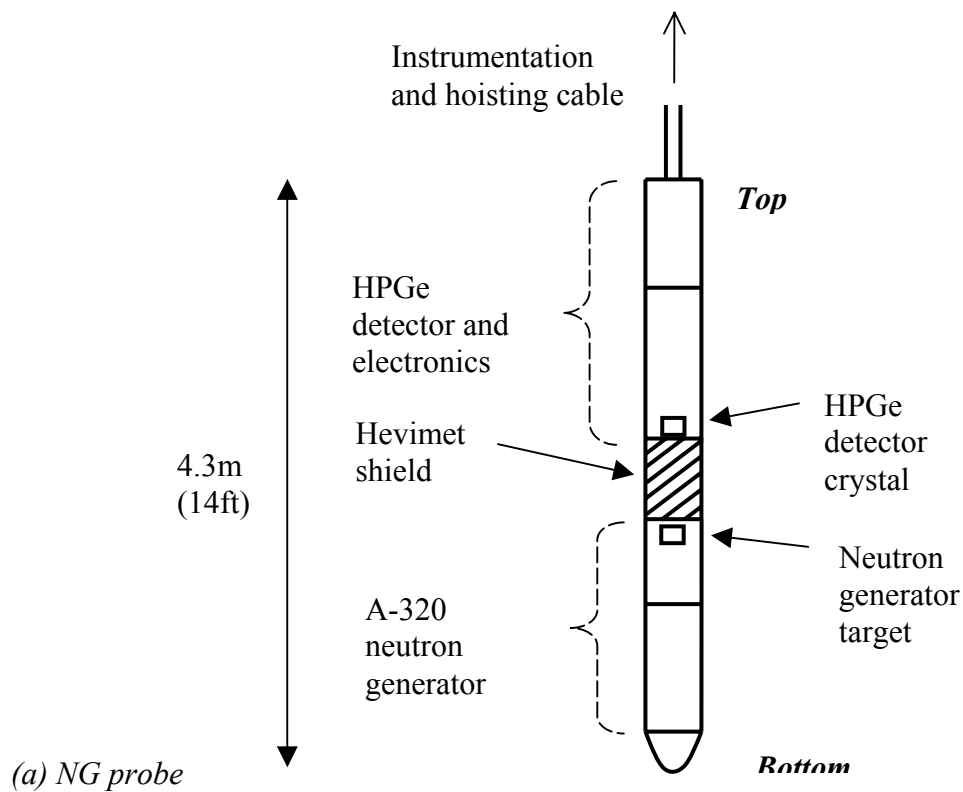


Figure 1: Assembly layout of the two probes. In the NG probe the two systems are separated by a Hevimet shield that attenuates both the neutron and gamma flux.

**Note:** Not to scale.

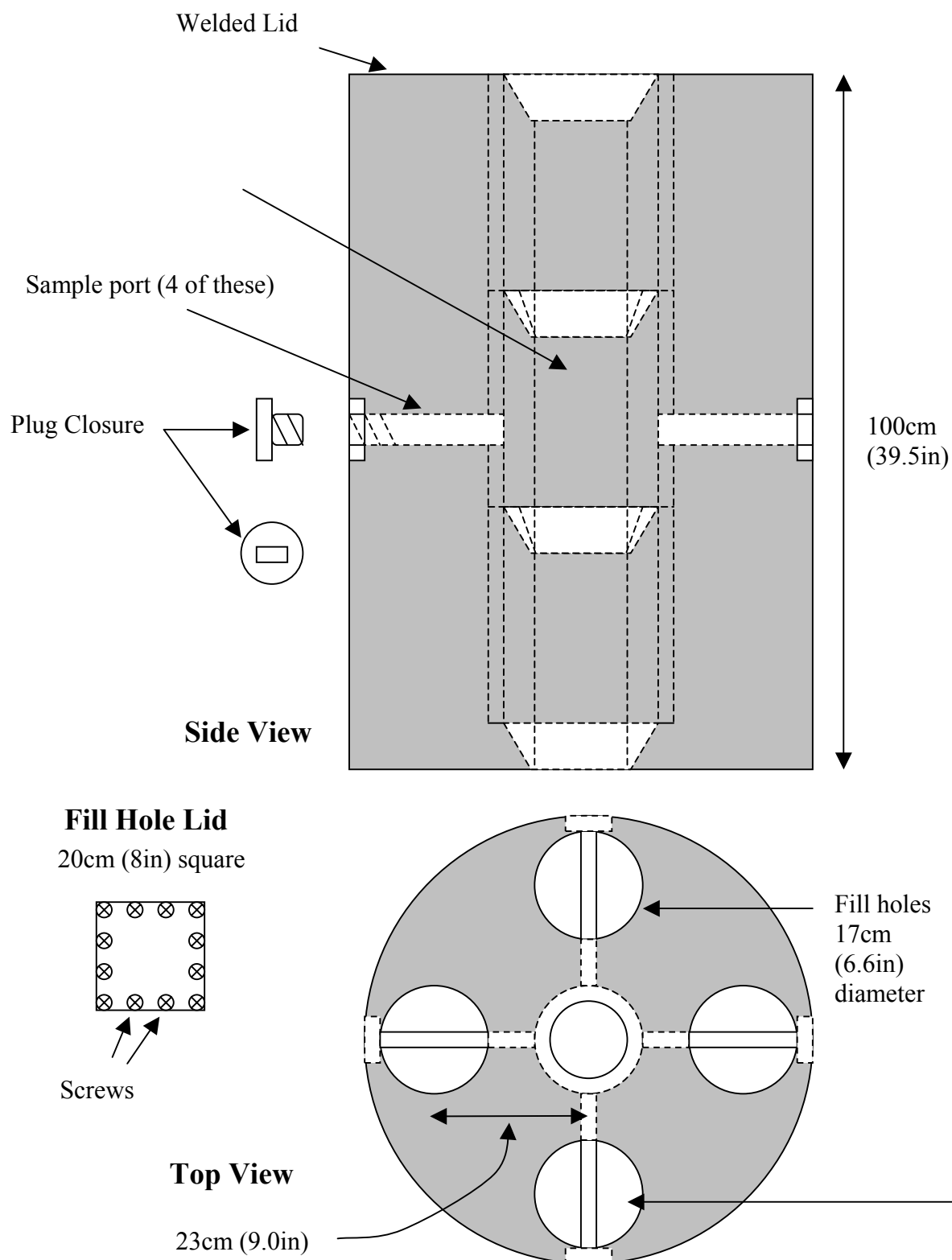


Figure 2: The design of an SLS. Each SLS is constructed from a modified 83gallon drum. Three pieces of penetrometer pipe run down along the longitudinal axis, while samples can be placed into the sample ports around the periphery of the surrogate. The surrogate is filled through the fill holes in the drum lid. The fill holes are to be covered when a surrogate is filled. **Note:** Not to scale.

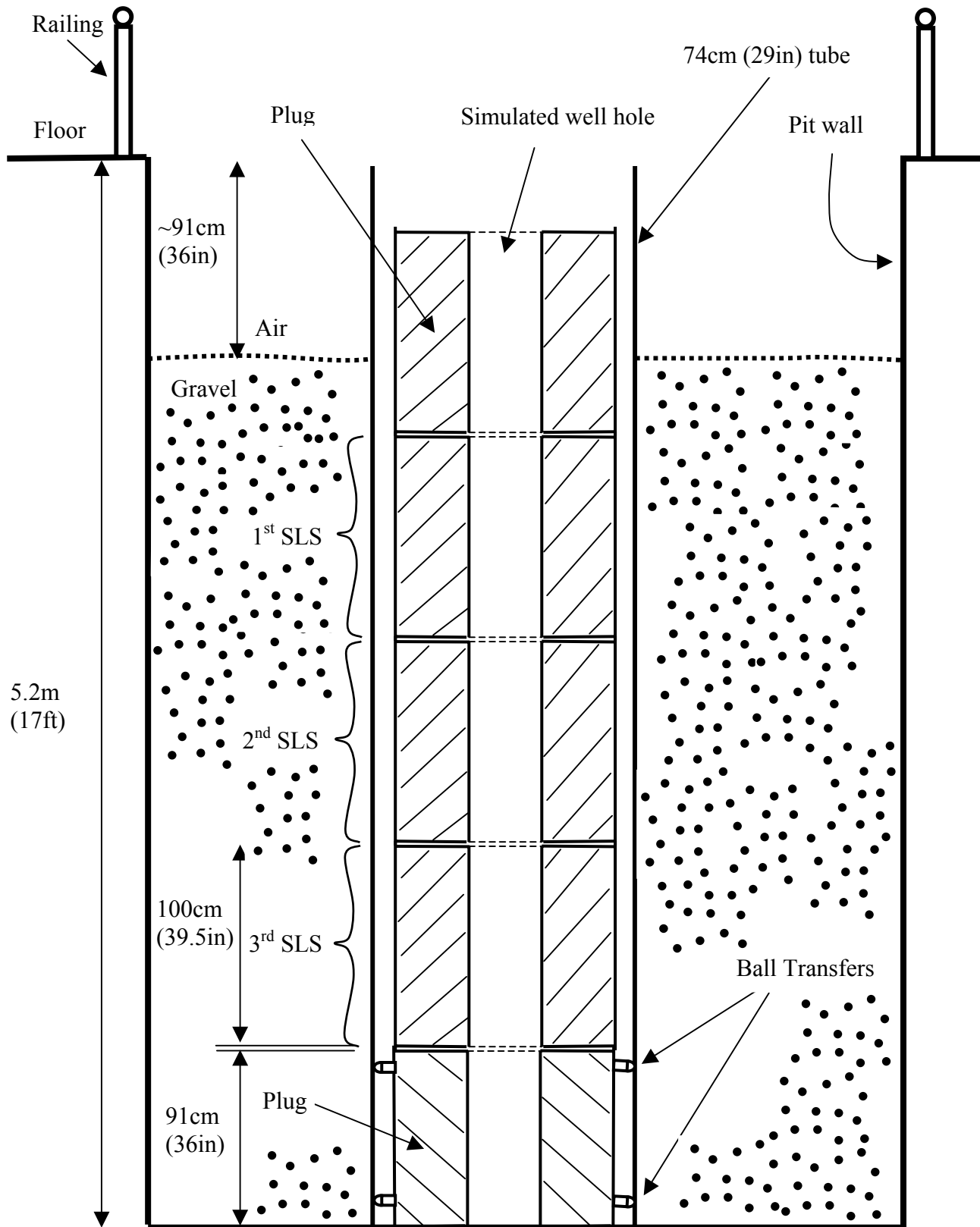


Figure 3: A stack of surrogates is shown placed within the 29in tube in the pit. All surrogates have the same dimensions. Plugs are shown at the top and bottom that are surrogates filled with concrete. **Note:** This diagram is not to scale.