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Upgrades to the Radiochemistry Analysis of Gas Samples (RAGS) Diagnostic at the National Ignition Facility

Donald Jedlovec, Kim Christensen, Carol Velsko, Bill Cassata, Wolfgang Stoeffl, Dawn Shaughnessy, John Lugten, Tony Golod, Warren Massey, Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, CA USA 94551-0808

ABSTRACT

The Radiochemical Analysis of Gaseous Samples (RAGS) diagnostic apparatus operates at the **National Ignition Facility** (NIF). At the NIF, xenon is injected into the target chamber as a tracer, used as an analyte in the NIF targets, and generated as a fission product from 14 MeV neutron fission of depleted uranium contained in the NIF hohlraum. Following a NIF shot, the RAGS apparatus used to collect the gas from the NIF target chamber and then to cryogenically fractionate xenon gas. Radio-xenon and other activation products are collected and counted via gamma spectrometry, with the results used to determine critical physics parameters including: capsule areal density, fuel-ablator mix, and nuclear cross sections.

We will discuss the following performance upgrades to the system:

Modification of the xenon fractionator to allow collection of krypton. Krypton is an important target analyte and is also generated as fission gas in NIF shots. We have modified the cryostat to enable fractionation of xenon, krypton, or both.

A translatable detector for quantification of the fission gases. An *in situ* Germanium detector is used to count the radio isotopes collected on the coldhead. NIF shots that incorporate depleted uranium in the hohlraum generate large amounts of fission gas, the radioactivity of which paralyzes the detector. We have designed a system that will adjust the detector-to-coldhead distance based upon either the count rate or the time after the NIF shot.

Keywords: krypton, radiochemistry, cryogenic, nuclear diagnostic

1. INTRODUCTION

This paper describes upgrades for two components of the system that are critical for the following applications of the RAGS diagnostic in the NIF:

Irradiation of a noble gas cell near target chamber center for cross section and other science experiments

Noble gas target implants (xenon, krypton), or target gas loads, for rho-r and collection efficiency measurements

Fission product yields

Mix studies with n,2n or charged particle reactions noble gas target implants

Accordingly, the system has been modified to fractionate, quantify, and extract krypton in the presence of milliCurie quantities of fission gas resulting from fission of uranium in the hohlraum in close proximity to the NIF target. This requires the operation at lower cryogenic temperatures. Also, the *in situ* detection system was modified so that the detector-to-coldhead distance could be remotely changed, based upon either the count rate or the time after the NIF shot.

A detailed functional description of the RAGS system has been provided by Shaughnessy¹. A brief summary follows.

RAGS is a modular design that cryogenically fractionates process gas from the NIF target chamber, isolating noble gases for transfer to in situ gamma detectors. In operation, the NIF target chamber cryopumps are isolated just before shot time

with the target chamber turbopumps alone delivering gas to the RAGS sampling line. The first module, a filter cart, contains a cold copper foam held at ~195K, two hot getters, and room temperature getters. It removes water vapor, hydrogen and most air gases, leaving primarily a noble gas and methane fraction. The collector cart fractionates noble gas on a cold head opposite an *in situ* germanium gamma detector. The waste stream is pumped to a reserve (abort) tank also equipped with a gamma detector. We also have the capability to heat to release a fractionated sample, collect it in a removable cold trap (sample bottle), and use the LLNL Nuclear Counting Facility germanium detector for decay counting to very accurately determine the amounts of radio isotopes collected. Figure 1 shows the complete RAGS instrument.

2. MODIFICATION OF THE XENON FRACTIONATOR TO ALLOW COLLECTION OF KRYPTON

The working component of the RAGS collector cart consists of a variable temperature cryogenic trap. The gas sampling line passes through a cryostat where a short section of the line, filled with copper foam, is cooled by a Gifford McMahon cryocooler. The foam-filled section is two inches long and is contained in a copper block. The block is cooled through a heat strap connected to the first stage of an Oxford/Austin Scientific Model 350, 2-stage cryocooler. The block also contains two RTD temperature sensors and four 25W cartridge heaters that allow the trap temperature to be measured and controlled. The sections of sampling line leading to and from the cryogenic trap consist of thin-walled stainless steel bellows that provide low thermal conductivity to the warm line and also accommodate thermal contraction. The gas sampling line enters and exits the cryostat through the same wall as the cryocooler, allowing easy assembly of the cryostat and excellent access to the contents when the cryostat is opened. Figure 2 shows the cryogenic trap assembly with the top of the cryostat removed. The cryogenic trap is located close to a thinned section of the cryostat wall that allows the *in situ* germanium detector to be placed outside the cryostat but very near the trap (Figure 3).

The RAGS collector cart was originally designed to fractionate xenon. The optimum trap temperature to achieve that was believed to be greater than 60 K. The original design achieves 60 K trap temperature with heaters off. Under these conditions, the heat load on the trap is dominated by the radiative heat load on the cold surface and the conduction load through the bellows in the gas sampling line. A 300 K blackbody radiation field produces a heat flux of 300 mW/in². Taking the cold surface area to be 110 in², and assuming the surface has 5% emissivity, the radiative load is approximately 1.63 W. The conductive heat flow through both stainless steel bellows is about 0.88 W, assuming for each bellows a heat flow path length of 5 inches and a wall thickness of 0.006 in. The cartridge heaters are powered by eight 26 AWG copper wires; the total heat flow through these wires is approximately 0.35 W. Gas flow through the sampling line is less than 10⁻⁵ g/s. The conductive heat flow through this gas is negligible, as is the heat required to cool it from 300K to 60 K. The total heat load on the first stage of the cryocooler is thus expected to be about 2.9 W. The load vs. temperature map for a Model 350 cryocooler shows a typical first stage temperature of about 44 K for this heat load. A 6061-T6 aluminum heat strap was designed to mechanically support the cold trap to the first stage of the cryocooler, and to sink 2.9 W from the trap when the trap temperature is 60 K and the cryocooler temperature is 44 K. In the original design, the measured trap temperature was in very good agreement with the temperature predicted by the thermal model. Under these conditions, the second stage of the cryocooler operates at about 11 K and serves to cryopump the cryostat.

With the desire to fractionate krypton, a new heat strap with identical geometry was fabricated from OFE copper. This strap carries about 3.0 W with its cryocooler end at 44 K and its trap end at 44.8 K. Thus the trap operates at about 45 K with no heater power applied. Again the measured performance was found to be in very good agreement with the thermal model. Initial operation has shown the ability to fractionate krypton. The copper heat strap provides a much tighter thermal coupling between the cryocooler and the trap, so more heater power is required to raise the trap temperature to release the fractionated species. Also, the second stage refrigerator temperature is increased by a larger amount when heating the trap compared to the original aluminum strap. Table 1 summarizes the thermal models for the trap with the aluminum and the copper heat straps when no heater power is applied.

Table 1 – Thermal model of the cryogenic trap as built to trap xenon and krypton

Xenon trap	Krypton trap
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Cold trap radiative heat load (W)	1.63	1.63
Bellows conductive heat load (W)	0.86	0.90
Heater wiring conductive heat load (W)	0.35	0.36
Heat load to cool carrier gas (W)	< 0.01	< 0.01
Gas conduction heat load (W)	< 0.06	< 0.06
Total cold trap heat load (W)	2.91	2.96
Cryocooler stage 1 temperature, from load/temperature map (K)	44	44
Measured cold trap temperature, no heater power (K)	60	45
Heat strap material	6061-T6 aluminum	OFE copper
Heat strap average conductivity (W/m/K)	63	1260
Heat strap thermal conductance (W/K)	0.19	3.7
Heat strap temperature drop (K)	16	0.8

3. A TRANSLATABLE DETECTOR FOR QUANTIFICATION OF THE COLLECTED FISSION GASES

The RAGS collector cart is equipped with a germanium detector in close proximity to the copper foam on which the noble gases (Xenon, Krypton) are collected. The geometry is shown in Figure 3.

The detector is a GEM Series High-Purity Germanium detector system with a ORTEC[®] DSPEC-50 gamma ray spectrometer. Data is acquired in list mode, as the requirement is to track in-growth and decay of fission gases that arrive at various times during the RAGS gas collection cycle.

On the collector cart, gas flows through, and is frozen onto, a 20% dense, porous insert of Duocel[®] copper foam (ERG Aerospace Corporation). This foam is in the form of a 125 cm² cube that is attached to the first stage of the cryopump. The external surfaces of the foam assembly do not communicate with the process vacuum but rather are internal to a secondary vacuum system that provides thermal isolation. The front face of the *in situ* detector is separated from the copper foam first by 2mm of copper, followed by a 1mm vacuum gap, then 1mm of steel, and finally an air gap to the detector of 6mm. Typically the *in situ* detector is used for process control measurements; the gas sample fractionated on the foam is ultimately heat-released to a secondary cold trap for extraction and analysis. While the detection geometry is not ideal, analytical results from the removable cold trap have established an accuracy of 10% for the amount of radioisotopes on the foam.

NIF target shots with D-T fuel and depleted uranium hohlraums generally have neutron yields in the range of 1E16. The resulting fast fission of the uranium can generate ~ milliCurie quantities of fission gas at the time of the shot. The total fission gas activity is down by two orders of magnitude after an hour and down three orders or more after six hours. However, without the ability to move the detector away from the source, dead time effectively blinds the detector. We have designed a system that adjusts the detector-to-coldhead distance, based upon either the count rate or the time after the NIF shot. The shot trigger initiates a programmed motion sequence; which is user-selectable. The translatable detector allows us to collect data on very short-lived radio-isotopes with the detector on the collector foam, and then to increase the distance for better measurements of longer-lived isotopes as the overall count rate increases due to the continual delivery of gaseous fission products to the system.

The translation stage from Linear Technologies[®] has a range of 20.0 inches and is controlled through a Galil[®] stepper motor controller. The Galil[®] stepper motor controller is managed from a rack-mounted diagnostic controller. The Galil[®] controller is pre-programmed with up to twenty pre-determined 'recipes' that allow the scientific staff to pre-select a distance and time where the detector can be moved anywhere in the 20 inch travel range. The movement of the stage is started by a shot trigger, which is provided locally by a DG645 pulse generator and timing module. Figure 7 is a system interconnect layout.

4. RESULTS

The additional radio-krypton gamma rays increased the count rate of the detector above the foam. In order to avoid detector paralysis and long recovery times at high initial count rates, the detector had been manually set to ~12" above the collection foam. The translatable stage now enables the detector to be positioned against the foam at shot time, maximizing the measurement of short-lived species with high activities present during first several minutes. As additional gas is pumped to the collection foam throughout the 15-minute collection interval, increasing the total count rate, the detector is then moved to 12" to permit counting of species with half-lives of several minutes or longer. Finally, after several hours, the detector is moved closer to the foam, for more efficient counting of fission products with half-lives greater than several hours. Subsequent temperature cycling of the copper foam is used to isolate xenon from krypton for sequential collection of the gases in bottles containing activated charcoal and cooled with liquid nitrogen.

When the cryogenic trap was at 60K, most of the krypton passed over it and into the abort tank. Figure 8a shows the krypton photopeaks for the gas in the abort tank. Once the fractionator modification reduced the base temperature to 44K, we used RAGS to collect gas on several shots that produced fission gases in order to characterize its performance. At this lower temperature, both radio-krypton and -xenon products are trapped completely on the copper foam, and no gamma rays from either of these elements has been detected in the abort tank (refer to Figure 8b).

5. ACKNOWLEDGEMENTS

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Figure 1. The Radiochemical Analysis of Gas Samples (RAGS) apparatus at the National Ignition Facility

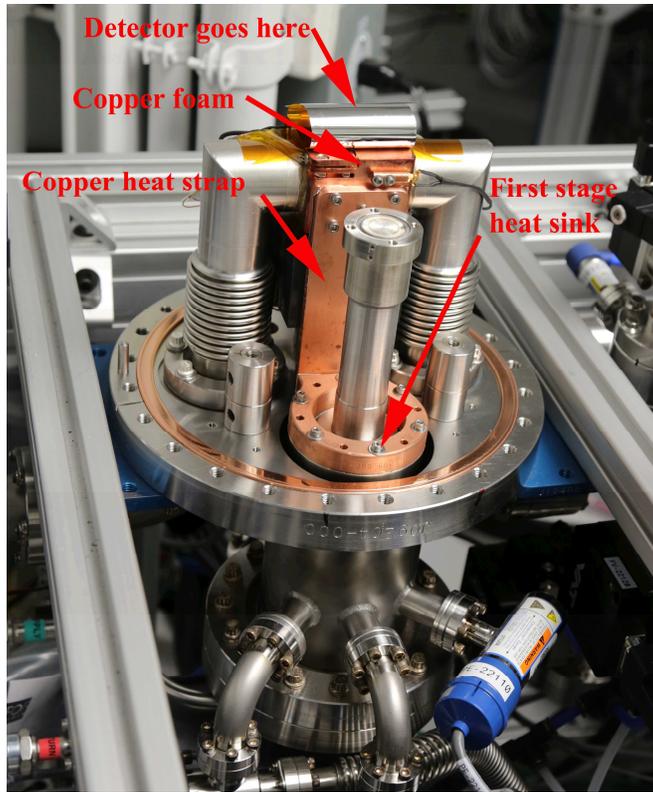


Figure 2. Coldhead, with vacuum housing removed

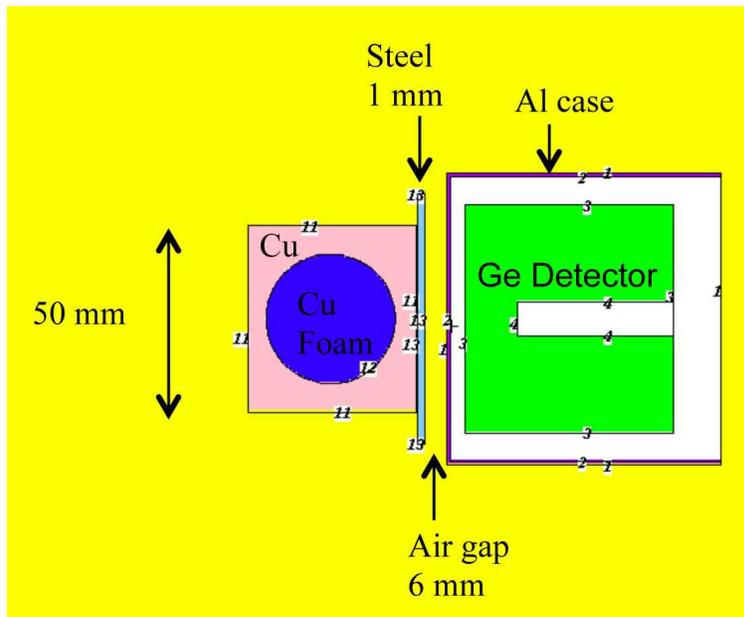


Figure 3. Detector geometry

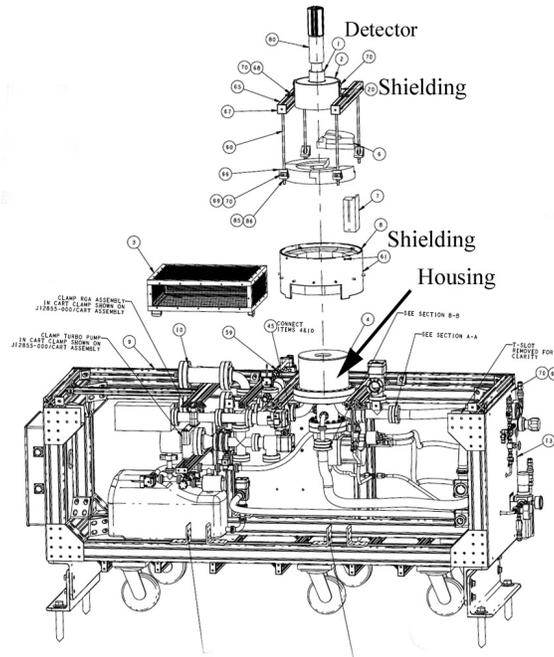


Figure 4. RAGS collector cart

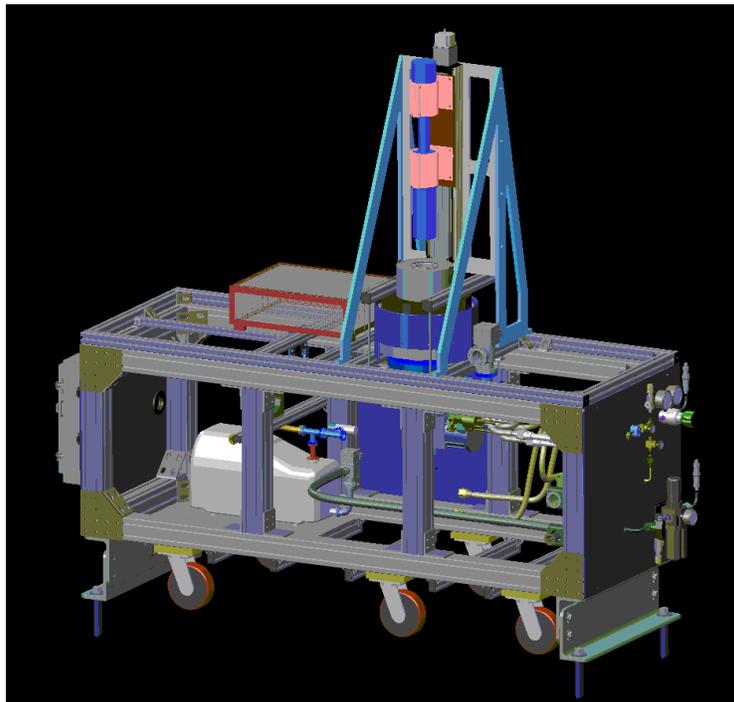


Figure 5. RAGS Collector Cart with Translatable Detector

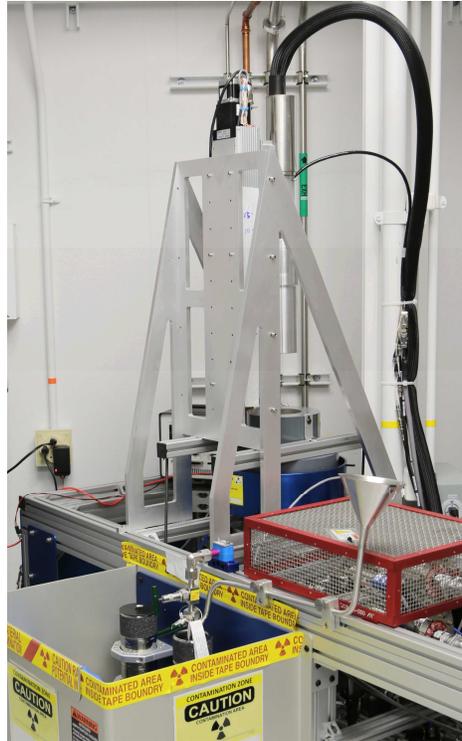


Figure 6. RAGS Collector Cart with translatable detector

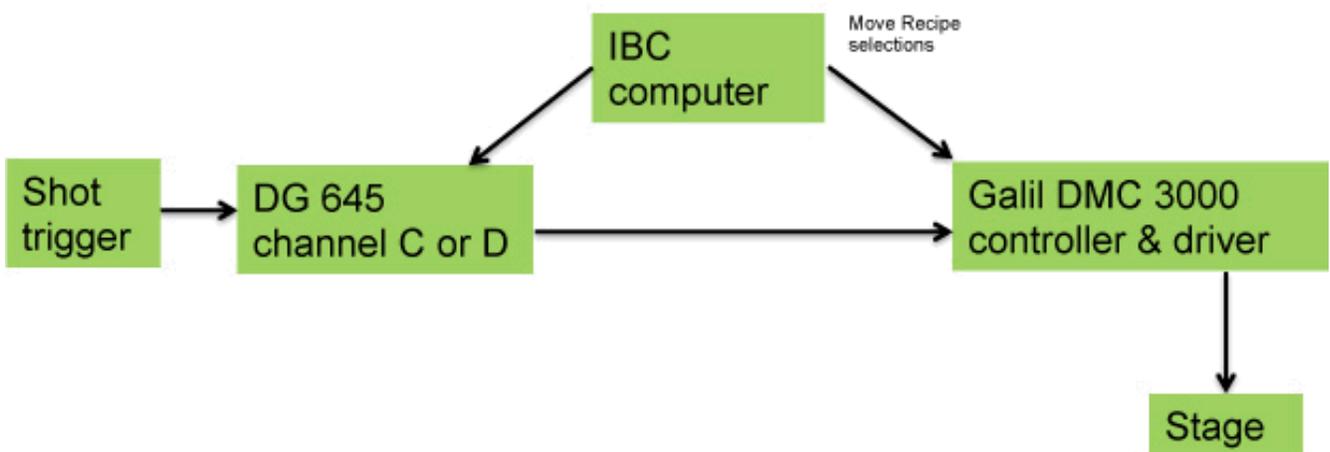
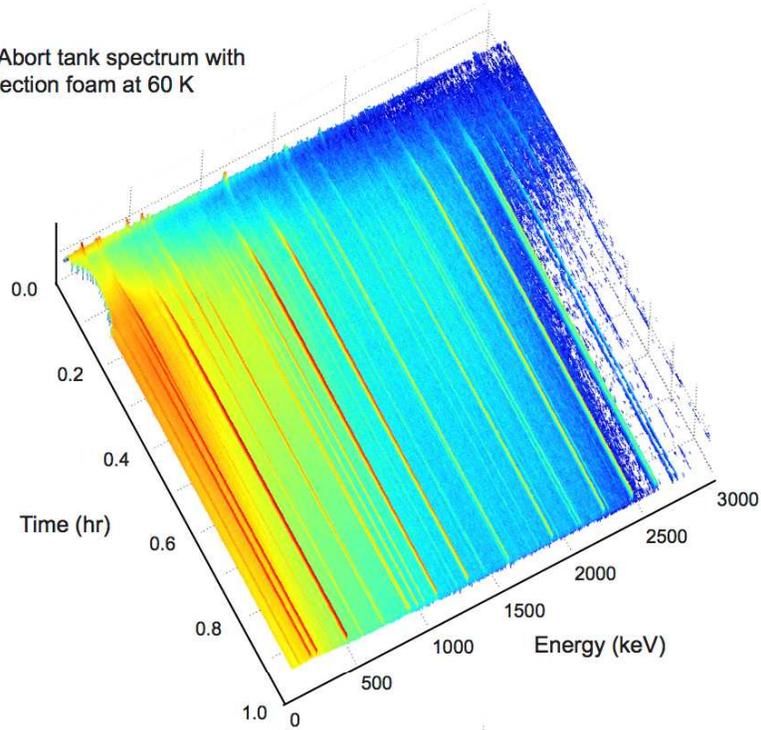


Figure 7. System Interconnect layout for the translatable detector

Color is proportional to the log of the count rate

a.) Abort tank spectrum with collection foam at 60 K



b.) Abort tank spectrum with collection foam at 44 K

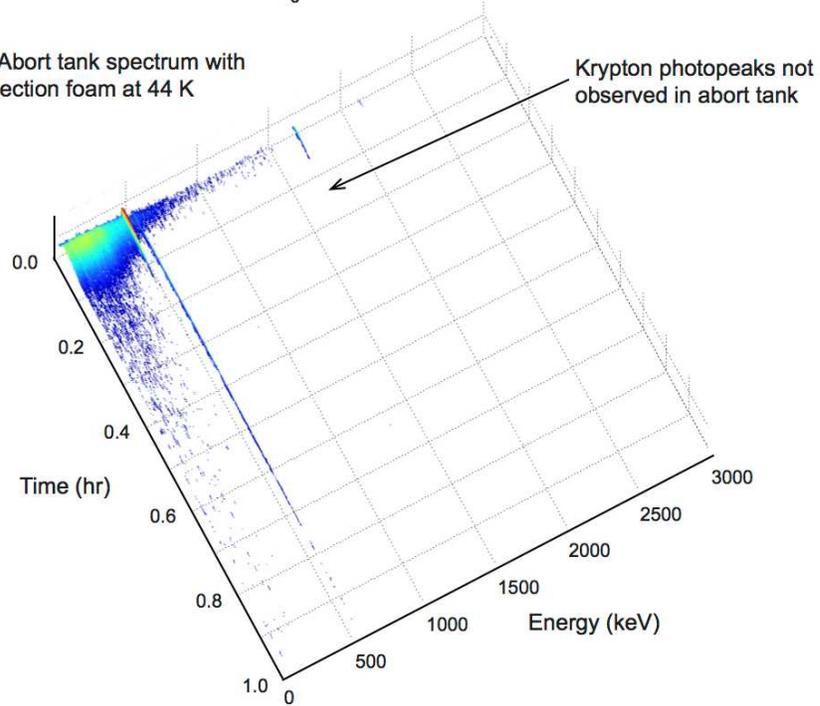


Figure 8. 3-D histograms showing effectiveness of krypton collection

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- [1] Shaughnessy, D. A., et al “the Radiochemical Analysis of Gaseous Samples (RAGS) apparatus for nuclear diagnostics at the National Ignition Facility, *Rev. Sci. Instrum.* **83**, 10D917 (2012); DOI: 10.1063/1.4742145.