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CTBTO Contractor Laboratory Test Sample Production Report

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ABSTRACT

In October 2012 scientists from both Idaho National Laboratory (INL) and the CTBTO contact laboratory at Seibersdorf, Austria designed a system and capability test to determine if the INL could produce and deliver a short lived radio xenon standard in time for the standard to be measured at the CTBTO contact laboratory at Seibersdorf, Austria. The test included sample standard transportation duration and potential country entrance delays at customs. On October 23, 2012 scientists at the Idaho National Laboratory (INL) prepared and shipped a Seibersdorf contract laboratory supplied cylinder. The canister contained 1.0 scc of gas that consisted of 70% xenon and 30% nitrogen by volume. The t_0 was October 24, 2012, 1200 ZULU. The xenon content was 0.70 +/- 0.01 scc at 0 °C. The ^{133m}Xe content was 4200 +/- 155 dpm per scc of stable xenon on t_0 (1 sigma uncertainty). The ^{133}Xe content was 19000 +/- 800 dpm per scc of stable xenon on t_0 (1 sigma uncertainty).

CONTENTS

FIGURES

TABLES

ACRONYMS

CTBTO	Comprehensive Test Ban Treaty Organization
INL	Idaho National Laboratory
scc	standard cubic centimeter at atmospheric pressure and 0 degrees centigrade

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1. INTRODUCTION

In October 2012 scientists from both Idaho National Laboratory (INL) and the CTBTO contact laboratory at Seibersdorf, Austria designed a system and capability test to determine if the INL could produce and deliver a short lived radio xenon standard in time for the standard to be measured at the CTBTO contact laboratory at Seibersdorf, Austria. The test included sample standard transportation duration and potential country entrance delays at customs. On October 23, 2012 scientists at the Idaho National Laboratory (INL) prepared and shipped a Seibersdorf contract laboratory supplied cylinder. The canister contained 1.0 scc of gas that consisted of 70% xenon and 30% nitrogen by volume. The t_0 was October 24, 2012, 1200 ZULU. The xenon content was 0.70 +/- 0.01 scc at 0 °C. The ^{133m}Xe content was 4200 +/- 155 dpm per scc of stable xenon on t_0 (1 sigma uncertainty). The ^{133}Xe content was 19000 +/- 800 dpm per scc of stable xenon on t_0 (1 sigma uncertainty).

1.1 LABORATORY ACTIVITIES

To produce this standard, expansion and mixing of gases was employed. Production included the mixing of xenon and nitrogen gases in a 70% / 30% (v/v) ratio. An 8 cc gas tight syringe was utilized for mixing. The gas manifold configured for this production run is shown in Figure 1.

In addition, Figure 1 displays the full outlet volume in orange, including transfer tubes, by which all xenon and nitrogen pressures were calculated. The full volume also includes the volume between the syringe plunger and the valve directly below the syringe. Table 1 displays manifold volumes as used in this production run.

Table 1. Manifold volumes used on the 10-23-2012 production

Section of Manifold	Volume (cc)
Inlet Volume - Includes volume of bulb 15-9	24.59
Needle Valve Open – Known volume metering valve	33.97
Valve#1 – Large dead-leg volume	14.90
Valve #2 – Small dead-leg volume	5.15
Valve #3 – Volume above syringe and below syringe plunger	1.66
Valve #4 – Tee fitting to first transfer tube	7.91
Transfer tube (1831)	3.46
Valve #5 – Cross fitting to second and third transfer tube	12.15
Transfer tube (1809)	3.48
Transfer tube (1835)	3.44
Valve #6 – Cross fitting to sample bulb and quick disconnect	9.18
Sample bulb (R16)	19.91

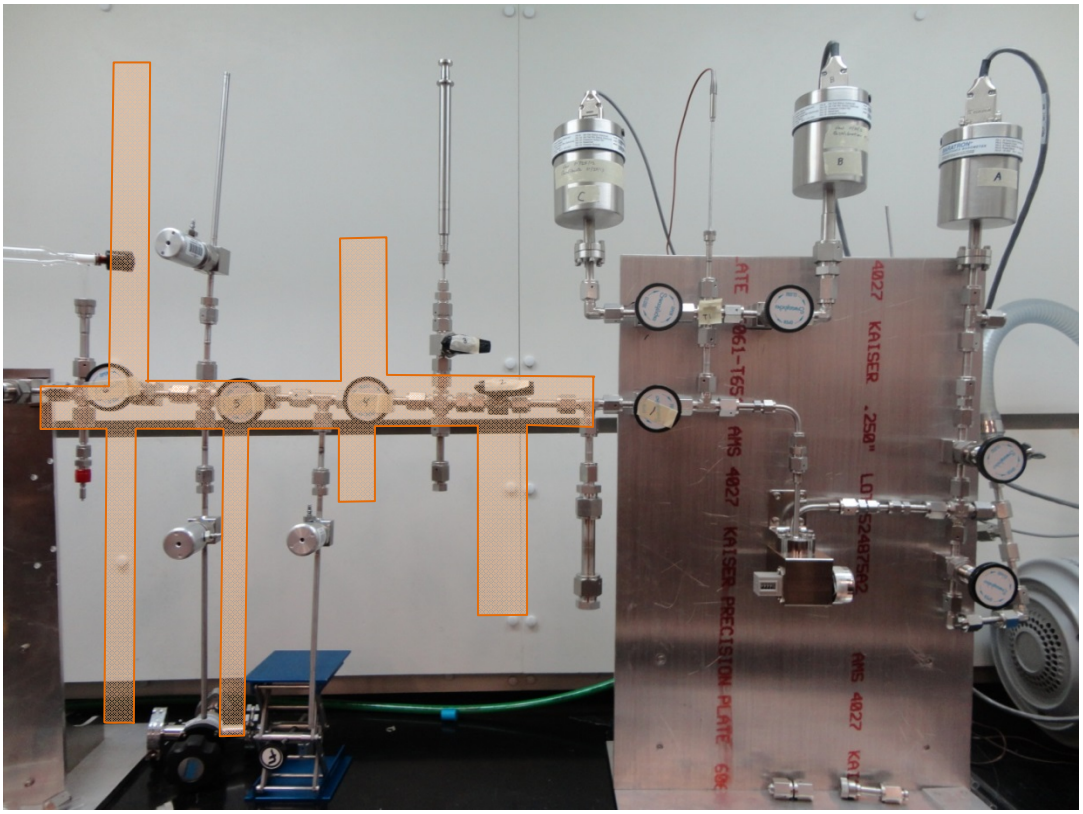

Figure 1. Manifold configured for the production run using an 8 cc gas tight syringe and full outlet volume shown in orange, including transfer tubes, by which all xenon and nitrogen pressures were calculated.

1.2 General Production Method

The following is a description of the general production method for this standard including a description of the mixing protocol.

Four freshly irradiated quartz vials containing both ^{133}Xe and ^{133m}Xe (~1 mL in size) were received from the University of Texas. The quartz vials had previously been filled with a nominal 0.5 cc of target ^{132}Xe at the INL and sent to Texas for irradiation.

Upon receipt of the irradiated targets at INL, all target vials were assayed. Individual targets were placed in the bean crusher, broken and the gaseous contents cryogenically transferred to a glass bulb, where they were then assayed again. The bulb had been previously filled with ~3 cc of stable xenon to provide ample carrier gas for cryogenic transfer of the target material. Based on the activity of the bulb, a glass bean was then prepared to match the requested xenon spike activity. The spike bean was prepared via a known volume where pressure and temperature are measured and recorded. The known volume is shown in purple in Figure 2.

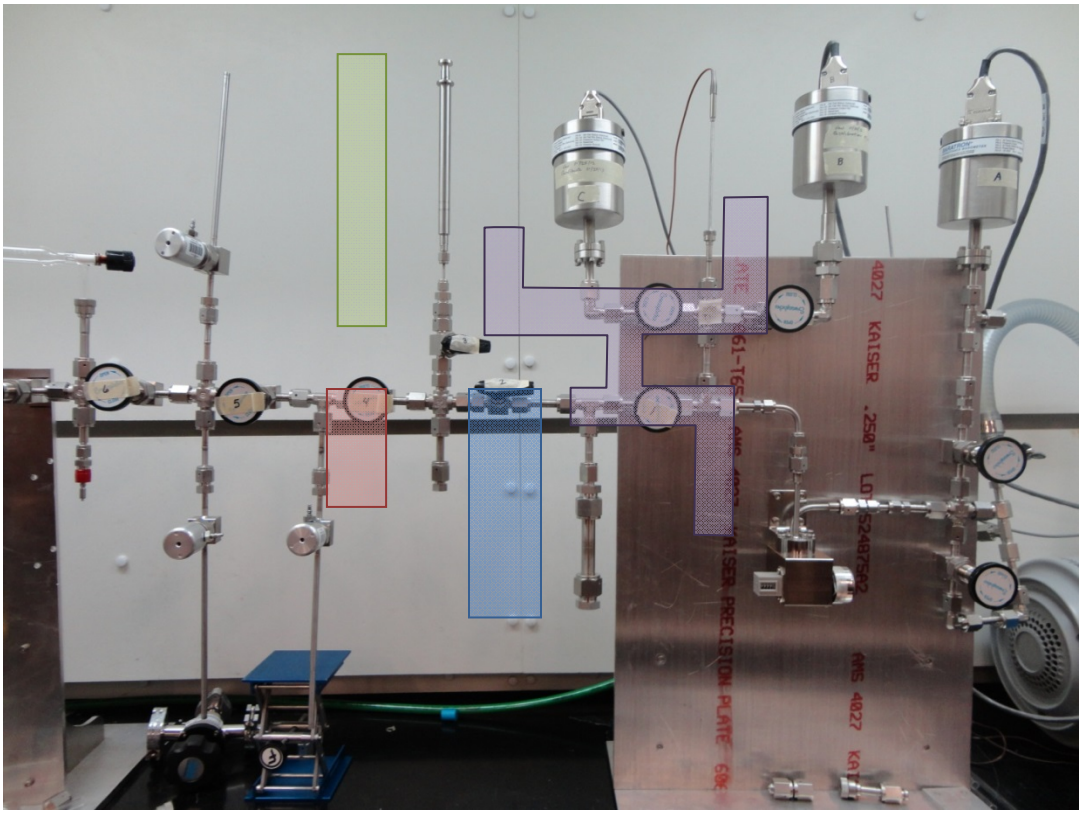

Figure 2. Manifold displaying known volume (purple), small dead-leg volume (red), large dead -leg volume (blue) and the 8 cc gas tight syringe (green).

Once a suitable spike bean is produced and the radio xenon quantified, the manifold is purged and evacuated, the spike bean is placed in the bean crusher, broken and the contents cryogenically transferred to the small dead-leg volume on the manifold. Figure 2 displays the small dead-leg volume in red as configured on the manifold. Once the spike transfer is complete, valves on either side of the small dead-leg are closed, isolating the xenon.

To meet the requested 70% xenon matrix gas, additional stable xenon is added by way of the known volume and cryogenically transferred to the small dead-leg volume. Valves on either side of the small dead-leg volume are closed following cryogenic transfer.

The total xenon added is the sum of the stable xenon present with the ^{133m}Xe spike bean and the stable xenon matrix gas. All gas is added through the known volume where pressure and temperature are recorded, followed by cryogenic transfer to the dead leg in the manifold (red area in Figure 2).

Table 2. Data from the production of ^{133m}Xe standard.

Data Needed for Calculations	Values
Activity of ^{133m}Xe in spike bean	175124 dpm
Activity of ^{133}Xe in spike bean	803000 dpm
Pressure in known volume transferred to the bean	45.56 torr
Working temperature	292.95 °K
Verification activity of ^{133m}Xe in the Schlenk tube	25421 dpm

Verification activity of ^{133}Xe in the Schlenk tube	101000 dpm
Volume of Schlenk tube	19.36 cc
Pressure of Schlenk Tube filled for verification	369.8
Volume of Canister	495.56 cc
T_0	October 24, 2012, 1200 Zulu

1.3 Xenon Calculations

Total transfer tube volume.

$$3.44cc + 3.46cc + 3.48cc = 10.38cc$$

Total volume of expansion area

$$14.90cc + 5.15cc + 10.38cc + 7.91cc + 12.15cc + 1.66cc = 52.15cc$$

Total pressure need in expansion area to provide 4 scc of gas in three transfer tubes at 292.95 °K.

$$\frac{(760torr)(12cc)(292.95°K)}{(10.38cc)(273.15°K)} = 942.3\ torr$$

Partial pressure of xenon in expansion area is 70% of the total thus

$$942.3 \times 0.70 = 659.6\ torr$$

Contribution of xenon in spike bean to pressure in the expansion volume

$$\frac{(45.56torr)(33.97cc)}{(52.15cc)} = 29.7torr$$

Additional xenon pressure needed in expansion volume to have total desired xenon.

$$659.6torr - 29.7torr = 629.9torr$$

Pressure of xenon needed in known volume

$$\frac{(629.9torr)(52.15cc)}{33.97cc} = 967.0\ torr$$

To prepare the final mixture, additional stable xenon required to dilute the radioxenon. The source of xenon was placed on the right side of the manifold shown in Figure 2. All volumes of the manifold were evacuated except the small dead leg holding the ^{133m}Xe spike. Then stable xenon was metered into the known volume (purple area Figure 2) until the pressure reached the equivalent of 967.0 torr at 0°C. The xenon spike in the dead leg was frozen by submersion of the dead leg in liquid nitrogen. Then, the valves between the known volume and the xenon dead leg (red area in Figure 2) were opened. This transferred all the stable xenon to the dead leg as observed by pressure measurement.

Prior to nitrogen addition, the known volume (by way of the needle valve) and inlet side of the manifold were evacuated. Nitrogen gas was expanded into the large dead-leg volume and known volume to provide ample pressure in the nitrogen dead leg (blue region in Figure 2) to meet the 30% nitrogen matrix gas. Following nitrogen addition, the valve between the large dead-leg volume and known volume was closed.

1.4 Nitrogen Calculations

Partial pressure of nitrogen in expansion area is 30% of the total thus

$$942.3\ torr \times 0.30 = 282.7\ torr$$

Pressure of nitrogen needed in the large dead leg volume

$$\frac{(282.7torr)(52.15cc)}{14.90cc} = 989.45\ torr$$

The large dead-leg volume was fabricated from a 1/4 in. female-to-female VCR union, 1/2 in. male VCR to 1/4 in. male VCR reducer, 1/2 in. female-to-female VCR union, and terminated with a 1/2 in. VCR plug. With the xenon and nitrogen matrixes contained within the small and large dead-leg volumes, the ball valve separating the two was then opened allowing for mixing.

1.5 Mixing Procedures

Production utilized an 8 cc, air tight, stainless steel syringe manufactured by Havard Apparatus. Mixing was conducted by manipulating the syringe piston to maximum and minimum volumes 50 times with a 5 second pause between each reverse in expansion or compression.

Once mixing of xenon and nitrogen gases was complete, valves isolating the transfer tubes from the gas mixture were opened and gas expanded. Transfer tubes farthest from the mixing zone were opened first followed by transfer tubes that were closer. Transfer tubes were then closed in the order they were opened. Transfer tubes were then assayed to verify the entire process.

To verify the activity, a 15-cc Schlenk tube was installed at the end of the manifold and filled with a portion of the remaining xenon/nitrogen gas by expansion. Pressure within the Schlenk tube was recorded and combined with the calculated volume of the full outlet volume (minus transfer tubes). Although the counting geometry of the Schlenk tube is not the best, it was chosen over the bean geometry as cryogenic transfer into a bean was not possible due to the nitrogen gas in the sample matrix.

1.6 ^{133m}Xe Activity and Activity Verification Calculations

Calculated ^{133m}Xe activity per scc of produced gas standard at 0°C on October 24, 2012, 1200 Zulu.

$$\frac{175124dpm}{52.15\ cc} \times \frac{760torr}{942.3torr} \times \frac{292.95°K}{273.15°\text{K}} = 2905\ \ dpm\ 133\text{mXe}/scc$$

Calculated ^{133m}Xe activity per scc xenon gas in standard blend at 0 °C on October 24, 2012, 1200 Zulu.

$$\frac{175124dpm}{52.15\ cc} \times \frac{760torr}{659.3torr} \times \frac{292.95°K}{273.15°\text{K}} = 4152\ \ dpm\ 133\text{mXe}/scc$$

Total scc in verification Schlenk tube at 0°C

$$19.36cc \times \frac{369.8torr}{760torr} \times \frac{273.15°K}{293.05°K} = 8.78\ scc$$

^{133m}Xe activity verification per scc of produced gas in verification Schlenk tube

$$\frac{25420.74\ dpm}{8.78scc} = 2895\frac{dpm}{scc}\ \ 133\text{mXe}\ on\ October\ 24, 2012, 1200\ ZULU$$

Activity of ^{133m}Xe per scc xenon in verification gas sample

$$\frac{2895\ dpm/scc}{0.70} = 4143\frac{dpm}{scc}\ on\ October\ 24, 2012, 1200\ zulu$$

1.7 ^{133}Xe Activity and Activity Verification Calculations

Calculated ^{133}Xe activity per scc of produced gas standard at 0°C on October 24, 2012, 1200 Zulu.

$$\frac{803000dpm}{52.15cc} \times \frac{760torr}{942.3torr} \times \frac{292.95°K}{273.15°K} = 13{,}300\ dpm\ 133\text{Xe}/scc$$

Calculated ^{133}Xe activity per scc of xenon gas in standard blend at 0°C on October 24, 2012, 1200 Zulu.

$$\frac{803000dpm}{52.15cc} \times \frac{760torr}{659.3torr} \times \frac{292.95°K}{273.15°K} = 19{,}000\ dpm\ 133\text{Xe}/scc$$

^{133}Xe activity verification per scc in verification Schlenk tube

$$\frac{101000dpm}{8.78\ scc} = 11500\frac{dpm}{scc}\ on\ October\ 24, 2012, 1200\ zulu$$

Activity of ^{133}Xe per scc xenon in verification gas sample

$$\frac{11500\ dpm/scc}{0.70} = 16400\frac{dpm}{scc}\ on\ October\ 24, 2012, 1200\ zulu$$

The verification geometry is the Schlenk tube, which was not well calibrated at the time of this measurement. There is good agreement between the ^{133m}Xe numbers by this method because the ^{133m}Xe gamma energy is from a less steep part of the energy efficiency curve. The ^{133}Xe verification is not as good but does fall within 2 sigma of the calculated value. The geometry of the Schlenk tube is part of the problem. The ^{133}Xe gamma energy is from a steep part of the energy efficiency curve and thus we would expect the lower validation count. INL will calibrate the Schlenk tube for ^{133}Xe when the next NIST ^{133}Xe becomes available.

In addition to the verification Schlenk tube, a CTBTO canister was installed at the end of the manifold. The CTBTO provided canister was filled with 1.0 scc of xenon/nitrogen gas mixture. Gas transfer was completed by evacuating the known volume on the feed side of the manifold. The needle valve was then opened allowing for gas flow in the reverse direction, reducing the production side of the manifold to the calculated target pressure to provide the 1.0 scc in the CTBTO provided container.

Pressure of gas to contain 1 scc of total gas at 0°C in the CTBTO provided container.

$$760torr \times \frac{1cc}{495.56cc} \times \frac{292.95°K}{273.15°K} = 1.65torr$$

Table 3 Physical constants used

Isotope	Half Life (units)	Energy (keV)	Branching Ratio (%)
^{131m} Xe	11.84 days	163.93	1.95
^{133} Xe	5.243 days	80.997	38
^{133m} Xe	2.19 days	232.221	10
^{135} Xe	9.14 hours	249.794	90