Big Xe sample technology

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INTRODUCTION AND MAIN RESULTS

At IMS stations, the xenon (Xe) sample volume usually does not exceed a few cubic centimeters, and for OSI samples it is even smaller. The proposed technology increases the Xe sample volume by more than 100 times with possibility to perform highly sensitive spectrometric measurements. The minimum detectable concentrations (MDCs) for Xe-133, Xe-135, and Xe-133m are below 10^{-5} Bq/m^3, which is several orders of magnitude lower than the MDCs of existing IMS Xe systems. This poster presents the principle of the large-volume sample technology and demonstrates how it can significantly enhance the sensitivity of Xe equipment and strengthen the treaty verification regime.

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Introduction

Noble Gas (NG) technology plays a key role in the CTBTO verification regime. Since the beginning of the International Noble Gas Experiment (INGE), NG equipment developed by different teams and institutions has evolved from prototypes into sophisticated, mature instruments, now deployed at various IMS facilities. Today, this equipment has nearly reached its sensitivity limit (MDC). **However, this sensitivity remains insufficient for the verification regime.**

Current IMS and OSI xenon systems cannot reliably measure all four Xe isotopes (Xe-133, Xe-135, Xe-133m, and Xe-131m) at their background levels. In most cases, only Xe-133 is detected. Furthermore, IMS and OSI systems are unable to consistently determine fluctuations of all four isotopes above background after nuclear tests. For example, spectra collected after the DPRK tests did not provide conclusive evidence for all 4 Xe isotopes. To improve sensitivity, two key factors must be considered: the volume of xenon in the sample and the minimum detectable activity (MDA) of the detector system. The MDA of existing Xe systems is already close to the theoretical limit, leaving little room for improvement. The volume of air processed during sampling is also constrained by system design and IMS station infrastructure, making it impossible to scale up the Xe sample volume significantly.

At present, only the Big **Xe Sample Technology** offers a realistic path to further strengthening the treaty verification regime.

Main principle of the Big Xe Sample Technology

The key principle of the big sample technology is the preparation of a spectrometric sample containing more than 100cc of pure xenon, followed by spectrometric measurement with an MDA comparable with the current IMS Xe systems. During the method testing, the MDC values for the four Xe isotopes were several orders of magnitude lower than those of existing IMS Xe systems.

However, the infrastructure of radionuclide monitoring stations is not sufficient for installing such sampling units. The same applies to laboratory setups. Therefore, the technology relies on krypton-xenon concentrates produced by air separation plants, or on liquid oxygen samples obtained from oxygen plants. Obtaining a large xenon sample at specific locations is not a problem. Within a 400 km range, one can typically find a steel factory, an oxygen plant (often located near hospitals), or a large air separation facility. The figure below illustrates the locations of steel factories in the USA and Canada.

Big Xe sample from Xe-Kr concentrate

Preparation of the Xe spectrometric sample includes the following steps:

- Extraction of Xe–Kr concentrate produced by air separation plants (ASP) (≈92% Kr, 7% Xe, traces of Rn). We have studied the production process in detail and can determine the sampling start and stop times with a resolution of 6 hours. This parameter is critical for accurate MDC calculation.
- Enrichment of the Xe fraction and removal of Rn. Separation of the sample into pure Xe and Kr. (The Kr fraction may also be of interest for IAEA applications, and a dedicated detector is available for this purpose.)

During equipment testing, we successfully produced more than 1 liter of pure Xe from Xe–Kr concentrate, suitable for spectrometric measurements. Currently, more than 100 large-scale ASP facilities operate worldwide, and the cost of Xe–Kr concentrate is below 10 USD per liter.

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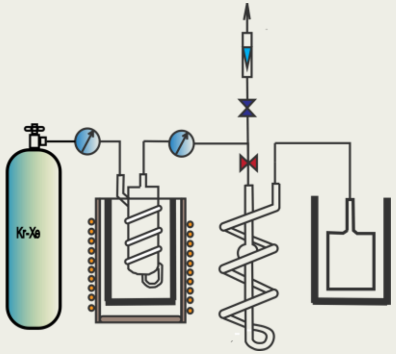
Separation unit for Xe-Kr concentrate

All technological processes for Xe sample preparation are carried out within a single installation that has a robust design and is easy to operate. The unit is portable and can be transported by vehicle or even packed into a few suitcases for air transport. Setup and testing take less than one hour. The system removes radon by a factor of more than 10^7 and eliminates residual krypton. The final spectrometric xenon sample can be collected in a syringe, a special ampoule, or a spectrometric cell, depending on whether the measurement will be performed on-site or in a laboratory. An aliquot of the Xe sample is taken for chromatographic analysis. The full working cycle lasts approximately 4 hours, depending on sample volume. The installation can produce up to 200 ml of pure Xe per sample, or about 1 liter of pure Xe per day.

Large Sample from Liquid Oxygen

Preparation of the Xe spectrometric sample includes the following steps:

- Sampling of 1 liter of liquid oxygen from an oxygen plant (initial Xe enrichment factor at factory sample ≈100). The time resolution of the sample is about 3 hours.
- Controlled evaporation of oxygen to further enrich Xe by a factor of up to 1000. Safety requirement: the concentration of acetylene must be well below 0.3 mg/dm³ to prevent explosion.
- Conversion of the remaining sample from liquid into gas phase.



- Final purification of the Xe sample from Rn, O_2, and N_2 using the same separation unit as for Xe–Kr concentrate.

The full preparation cycle lasts about 8 hours. From 1 liter of liquid oxygen, the installation produces approximately 50 cc of pure Xe suitable for spectrometric measurements.

History of the Large Xe sample technology

The technology was first developed by Yu. Popov and V. Prelovsky more than 50 years ago. Between the 1970s and the early 1990s, more than 5,000 samples were produced using this approach. At that time, large xenon samples were necessary for detection Xe-135, Xe-133m and Xe-131m in spectra. Only single NaI(Tl) detector was used for spectrometric measurements, and their MDA was significantly worse compared to modern Xe coincidence detector systems. Therefore, only large-volume of Xe in the sample gave chance to identify different isotopes reliably.
Today, our advanced coincidence detector systems with large measurement cell and high-resolution beta (SiPIN) detectors, allows not only to measure Xe-133, Xe-135, Xe-133m, and Xe-131m in each sample, but also to determine their ratios.
This demonstrates that the new big sample technology is, in fact, a revival and modernization of a once-forgotten method.

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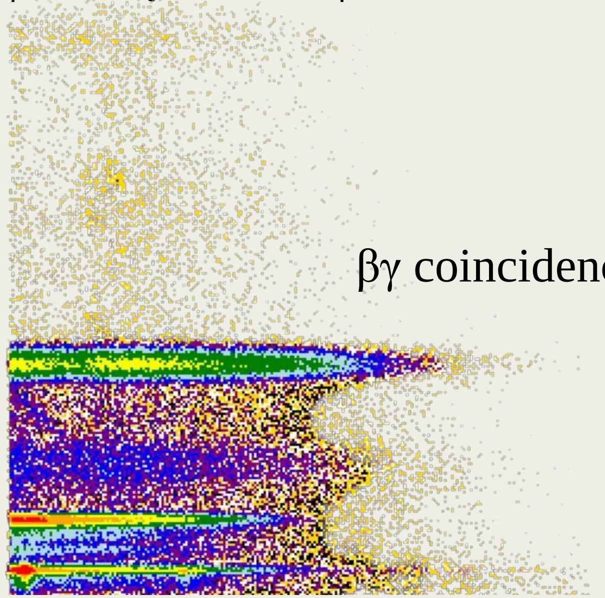
Detector System

The main feature of the detector system, operated in beta–gamma coincidence mode, is the large volume of its measurement cell. The latest version of the cell has a volume of 51 cc. The system consists of one NaI(Tl) gamma detector with a well (Ø37 mm × 50 mm) and a matrix of 12 SiPIN detectors arranged in a cylindrical cell. The system operates properly at pressures up to 2.5 atm. To reduce the free path of electrons, a cellular detector structure was implemented. The system has a very low memory effect (<0.04%). The beta detector resolution for Xe-131m is 10 keV, which makes it possible not only to separate Xe-131m from Xe-133m, but also to measure Xe-135 and from Xe-133m using only the beta spectra. Testing has shown that the MDC for Xe-133, Xe-135, Xe-133m, and Xe-131m is consistently below **10^{-5} Bq/m^3**

βγ detector assembly

New Software for the Detector System

The spectra always contain sufficient amounts of Xe^{133} for quality control; therefore, additional QC measurements, as performed in IMS NG equipment, are not required. Moreover, the large counting statistics of Xe in the sample spectra, combined with deep removal of radon, significantly reduce the measurement time for gas background. Currently, the duration of the gas background measurement is about 1 hour and sample measurement - 12h accordingly. For quality control purposes, we have implemented a neural network to monitor resolution, efficiency, and energy calibration. At present, QC software operates in test mode.



βγ coincidence histogram

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Improving the Treaty verification regime with Big Xe Sample Technology

Deployment of additional sampling points or temporary replacement of IMS stations in case of malfunction.

• Under a call-off contract with air separation or oxygen plants, Xe–Kr concentrate or LO samples can be delivered within one day after a PTS request.
• Several sets of mobile Xe equipment are stored at CTBTO headquarters. The equipment can be transported worldwide within a few days as regular luggage on commercial flights.
• The total weight of the installation (excluding the lead shield) does not exceed 50 kg.

Thus, it is possible to establish additional Xe monitoring sites around the globe within a very short time. If commission to assume violation of the Treaty this new monitoring points by an order increase chance to determine it.

Xe background study

With this technology, it is possible to study xenon background worldwide for all four CTBT-relevant isotopes, not only Xe^{133}, which is usually the only one reliably detected at present. Due to the large sample volume and high counting statistics, all four isotopes (Xe-133, Xe-135, Xe-133m, and Xe-131m) can be detected with negligible probability of false positives or false negatives.

The technology makes it possible to analyze the ratios between Xe-133, Xe-135, Xe-133m, and Xe-131m in background concentrations.

Big Sample Technology for OSI Tasks

During the OSI preparation phase and throughout the inspection, Xe measurements can be obtained from Xe–Kr concentrate or LO samples collected at facilities in neighboring countries or in the inspected country itself.

The large-sample technology makes it possible to begin measurements immediately after an explosion (including from neighboring countries), with a sensitivity far exceeding that of the current OSI NG equipment.

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