



Radioxenon spiked air



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ABSTRACT

Four of the radioactive xenon isotopes (^{131m}Xe , ^{133m}Xe , ^{133}Xe and ^{135}Xe) with half-lives ranging from 9 h to 12 days are produced from nuclear fission and can be detected from days to weeks following their production and release. Being inert gases, they are readily transported through the atmosphere. Sources for release of radioactive xenon isotopes include operating nuclear reactors via leaks in fuel rods, medical isotope production facilities, and nuclear weapons' detonations. They are not normally released from fuel reprocessing due to the short half-lives. The Comprehensive Nuclear-Test-Ban Treaty has led to creation of the International Monitoring System. The International Monitoring System, when fully implemented, will consist of one component with 40 stations monitoring radioactive xenon around the globe. Monitoring these radioactive xenon isotopes is important to the Comprehensive Nuclear-Test-Ban Treaty in determining whether a seismically detected event is or is not a nuclear detonation. A variety of radioactive xenon quality control check standards, quantitatively spiked into various gas matrices, could be used to demonstrate that these stations are operating on the same basis in order to bolster defensibility of data across the International Monitoring System. This paper focuses on Idaho National Laboratory's capability to produce three of the xenon isotopes in pure form and the use of the four xenon isotopes in various combinations to produce radioactive xenon spiked air samples that could be subsequently distributed to participating facilities.

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1. Introduction

A system to monitor Comprehensive Test-Ban-Treaty compliance under the Preparatory Commission is on a well-established path to maturity (Auer and Prior, 2014). This includes a planned network of 40 radioactive xenon monitoring stations with 30 stations that are established and operating (Auer and Prior, 2014; Auer et al., 2010; Ringbom et al., 2013). One component of this network is to differentiate a nuclear weapon detonation from nuclear power plant and medical isotope operations (Auer and Prior, 2014; Auer et al., 2010, 2004) using detection and ratio measurements of four radioactive xenon isotopes. The stations are constantly collecting samples and analyzing for radioactive xenon isotopes and isomers. These analyses consist of detection of the concentration in air and isotope/isomer ratios for each of the four isotopes and isomers: ^{131m}Xe , ^{133m}Xe , ^{133}Xe , and ^{135}Xe . When a seismic event is detected that has the possibility of being a nuclear test and the time

window for xenon production overlaps with the time of the seismic event, there is a high probability that the two correlate.

In support of the world-wide non-proliferation community, Idaho National Laboratory has developed a quality assurance and quality control material production capability for radioactive xenon. The two longer-lived isotopes (^{131m}Xe and ^{133}Xe) were the first to be isolated in pure form for use in standards because they are by far the easiest to procure, calibrate, and ship while still having adequate activity for accurate measurements. Addition of the other two isotopes/isomers with shorter half-lives (^{133m}Xe and ^{135}Xe) has occurred more recently.

These standards include pure individual isotopes/isomers for photon energy quality control, quantitative blends of radioactive xenon isotopes/isomers for isotope ratio measurements, and production of blended radioactive xenon in an air matrix for verification of the ability to quantify radioactive xenon concentrations collected from the atmosphere.

When collecting a sample from the atmosphere, it is necessary to obtain an accurate measurement of the total air volume in order to quantify the xenon activity concentration (Auer et al., 2010, 2004). This is accomplished by cryogenically trapping both

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natural and fission product xenon. A huge volume of air passes through the collection system with only the xenon component and a few impurities captured. There are three systems for this function in the International Monitoring System: ARIX (Dubasov et al., 2005), SAUNA (Ringbom et al., 2003), and SPALAX (Fontaine et al., 2004). The reader is referred to these references for the separation techniques. Once sample collection is finished, this condensate is analyzed for both radioactive xenon and total xenon volume (stable and radioactive). The total volume of air is obtained by measuring the total volume of xenon combined with the concentration of xenon in air. Knowing the total volume of air and the activity level of each of the radioactive xenon isotopes, the concentration of each radioactive xenon isotope in the air sample is calculated. Because stable xenon is used to determine the quantity of air, the ideal quality control and assurance materials must be prepared in air with little, if any, additional stable xenon. When there is no alternative to the addition of non-radioactive xenon, the total quantity must be known, passed on to users, and included in calculations. The verification standards must be well blended and characterized for activity and with sufficient volume to produce a single quality control sample to be sub-sampled and made available to all participating laboratories.

2. Radioactive xenon production

Several technologies are available for production of both pure and mixed radioactive xenon isotopes that can be subsequently incorporated into radioactive xenon spiked samples.

1. Irradiation of natural xenon to obtain a mixture of all radioactive xenon isotopes in a natural xenon matrix. This material has had limited applications because the ratios of the radioactive isotopes have been less than ideal for the intended applications.
2. Irradiation of a commercially available, mass separated, stable xenon isotope to obtain a single radioactive xenon isotope in a matrix of the mass-separated stable isotope (Haas et al., 2009). This can be used to produce radioactively pure ^{135}Xe (9.14 h) from mass-separated ^{134}Xe (stable). This is used to calibrate the gamma spectrum in the vicinity of the 249.8-keV photon as well as for beta–gamma coincidence systems. Because it contains stable ^{134}Xe at a level that influences measurement of the total volume of air, this must be included in that calculation.
3. Isolation of mixed radioactive xenon from fission products followed by isotope separation. This is used to obtain ^{133}Xe (5.2475d) that is subsequently mass separated from accompanying $^{131\text{m}}\text{Xe}$ and any other xenon isotopes. It is important to maintain very low levels of $^{131\text{m}}\text{Xe}$ because the difference in half-lives will allow the $^{131\text{m}}\text{Xe}/^{133}\text{Xe}$ ratio to increase over the course of time. ^{133}Xe can be used to calibrate the gamma spectrum in the vicinity of the 80.99-keV photon as well as for beta–gamma coincidence systems.
4. Production of pure $^{133\text{m}}\text{Xe}$ (2.198d) has been demonstrated (Perajarvi et al., 2010); however, with its short half-life, it constantly decays to the longer lived ^{133}Xe isomer. Thus, $^{133\text{m}}\text{Xe}$ cannot be incorporated into a pure standard that can be shipped to another location. It is useful at the location of production, but with any shipping involved, the result is a mixture of ^{133}Xe and $^{133\text{m}}\text{Xe}$. $^{133\text{m}}\text{Xe}$ could be used for a quality control check of the gamma spectrum in the vicinity of the 233.2-keV photon as well as for beta–gamma coincidence systems.
5. Chemical isolation of a single radioactive isotope of an element other than xenon that subsequently decays to a pure radioactive xenon isotope. ^{131}I is commercially procured and is “milked” to obtain pure $^{131\text{m}}\text{Xe}$ (11.84d) for a quality control check of the

gamma spectrum in the vicinity of the 163.9-keV photon as well as for beta–gamma coincidence systems.

6. Mixtures of all four radioactive xenon isotopes obtained by “milking” a ^{252}Cf source (McGrath et al., 2013). Application of this material is advantageous because it produces a radioactive xenon mixture with the known ^{252}Cf fission ratios. This radioactive xenon can be incorporated in any matrix and does not contain significant stable xenon to perturb the natural xenon content when used to spike air. Currently, this is the only source for $^{133\text{m}}\text{Xe}$ and ^{135}Xe without additional stable xenon.
7. Quantitative blends of various combinations of $^{131\text{m}}\text{Xe}$, ^{133}Xe , and ^{135}Xe in air. Blends of these isotopes are required when evaluating possible spectral interferences and for calibrating the relative sensitivities of gamma spectrometers for the respective gamma energies. Accurate measurement of isotope ratios based on these sensitivities across the Comprehensive Test-Ban-Treaty complex is critical when determining if detection of radioactive xenon (Auer and Prior, 2014) is from a nuclear detonation.

3. Experimental

3.1. Pure isotope production

3.1.1. Pure $^{131\text{m}}\text{Xe}$

This is the easiest radioactive xenon isotope to produce, quantify, and distribute due to the longer half-life and ease of production in pure form without a stable xenon matrix. $^{131\text{m}}\text{Xe}$ is the decay product of ^{131}I . ^{131}I is the decay product of ^{131}Te . ^{131}Te is produced by neutron irradiation of ^{130}Te . Because ^{131}I (8.02d) is used for medical diagnostics, it is commercially available and can be “milked” to obtain $^{131\text{m}}\text{Xe}$. It is received in a basic solution in a sealed bottle. The capped vial is allowed to sit for 10–14 days. The maximum quantity of $^{131\text{m}}\text{Xe}$ contained in a sealed vial of ^{131}I is reached at 12 days post-sealing, corresponding to secular equilibrium. Greater than 90% of the maximum occurs between days 9 and 15 post-sealing the vial. The bottle is placed inside a chamber (Fig. 1) equipped with a helium feed and an outlet fed into a charcoal-filled tube cooled in liquid nitrogen. For collection of $^{131\text{m}}\text{Xe}$, the chamber is purged with helium while the exit through the charcoal is submerged in liquid nitrogen. A typical purge time is 2–3 h.

To chemically purify the $^{131\text{m}}\text{Xe}$ from any ^{131}I that may have reached the charcoal trap, it is passed through a gas chromatograph. The timing of the $^{131\text{m}}\text{Xe}$ elution is monitored with a sodium iodide detector connected to an audible output. When the $^{131\text{m}}\text{Xe}$ peak begins to elute, the outlet of the gas chromatograph is switched onto a liquid nitrogen-cooled charcoal trap to capture the purified $^{131\text{m}}\text{Xe}$.

3.1.2. Pure ^{133}Xe

This is the second simplest radioactive xenon isotope to produce, quantify, and distribute due to the approximate 5-day half-life. Like ^{131}I , ^{133}Xe is a medically useful isotope and available commercially. It is derived from uranium fission followed by dissolution and gas extraction. In order to keep this isotope mostly free from ^{131}Xe , the sample is irradiated for only a short period and chemically processed in a short period. Because the difference in half-lives of the predecessor iodine isotopes is 20.8 h versus 8.02 days, the 133/131 ratio is relatively large (how large depends on the timing of the irradiation and reprocessing). It is still necessary to conduct isotope enrichment because a highly purified sample is easier to quantify. The ^{133}Xe starting material is procured as the mixed radioactive xenon in carbon dioxide. It is processed through Idaho National Laboratory's xenon mass separator (Fig. 2) to obtain high-purity ^{133}Xe (Appelhans et al., 2005).

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