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Pacific Northwest
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Abstract

Pacific Northwest National Laboratory has developed an automated system for separating Xe from air and can detect the following radioxenon isotopes, ^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe. This report details the techniques used to generate the various radioxenon isotopes that are used for the calibration of the detector as well as other isotopes that have the potential to interfere with the fission produced radioxenon isotopes. Fission production is covered first using highly enriched uranium followed by a description and results from an experiment to produce radioxenon isotopes from neutron activation of ambient xenon.

Radioxenon Generator Using HEU

The use of ^{135}Xe is instrumental in the calibration of the ARSA beta-gamma viewer as there are no readily available sources of this isotope because of its short half-life (9.1 hours). In the past ^{135}Xe has been obtained from neutron activation of micro-gram quantities of ^{235}Uat a research reactor located on the Washington State University at Pullman, Washington. Because of shipping requirements it was not possible to obtain the fission products from the irradiated samples for two days after irradiation. To overcome both the time delay and the administrative planning and coordination we have developed a radioxenon generator (RXG) that can be irradiated with sealed neutron source here at our laboratory. The neutron irradiation time can be varied over a period of hours to several weeks and produces ^{133}Xe and ^{135}Xe along with several other noble gas isotopes with varying ratios determined by the irradiation time.

The RXG is composed of 10.1 grams of U_3O_8 (uranium oxide) powder, with the uranium being 95% ^{235}U. A diagram of the RXG is shown in figure 1-A. The uranium-oxide powder is double sealed in three-ply 3COM SBMF filter paper to prevent the oxide powder from being dispersed and to allow fission product gases to diffuse out. The aluminum containment bottle was chosen both for reasons of safety (quartz bottles may break if dropped), and low activation products. The plastic cap and septum provide a nearly gas tight seal to minimize leakage of the gaseous fission products. The gas samples are

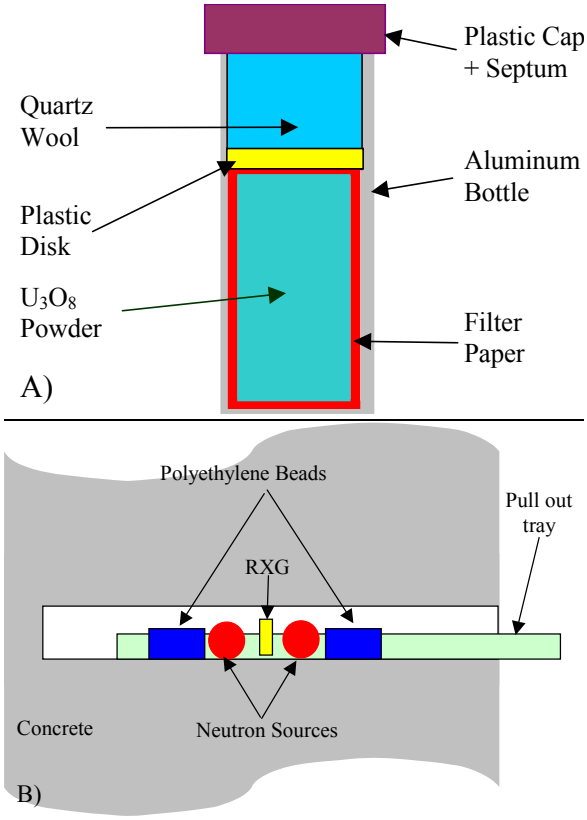

Figure 1-A) Schematic of the RXG. The overall length is 10 cm with a diameter of 2 cm. **1-B**) Neutron source configuration for irradiation of the RXG (not drawn to scale). The pull out tray is 5 meters long. The surrounding concrete is several meters thick.

drawn out of the RXG using a syringe with attached needle. To prevent the needle from penetrating the filter paper, and thus causing the unwanted spread of fission products, a loose fitting plastic disk is placed on the top of the oxide powder. Quartz wool is used to keep the disk and double bagged uranium-oxide powder from moving.

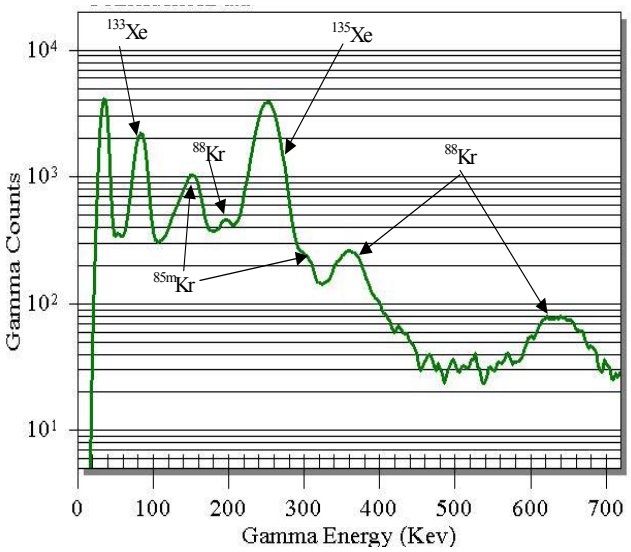

Figure 2. Beta-gated gamma spectrum of radioxenon and radio-krypton gases produced in the RXG. The peak at 30 keV contains several x-ray and gammas from several of the isotopes. Most of the strength for this peak (~90 %) is from ^{133}Xe followed by ^{135}Xe (~6%).

To irradiate the uranium-oxide powder, two neutron sources were used with moderating material. During the irradiation of the sample an 80 gram PuBe source with a neutron flux of 8.8 x 10^8 neutrons/sec and a 10 microgram ^{252}Ca source with a neutron flux of 2.8 x 10^7 neutrons/sec are placed on either sides of the RXG (see figure 1-B). To facilitate an increase in fission reactions, two plastic bottles filled with 385 grams of polyethylene beads are placed on either side of the sources as moderators to increase the flux of thermal neutrons. After a 2 week irradiation time there is approximately 25 mBq of ^{133}Xe, 50 Bq of ^{135}Xe, 10 Bq of ^{85m}Kr and 0.6 Bq of ^{88}Kr. From simplistic fission yield and neutron-flux rate calculations these values should be 3 times higher and it is currently believed that the uranium-oxide powered is holding up most of the fission gases.

To increase the yield produced by the RXG it may be advisable to use uranium-sterate, which allows emanation of fission gases more easily. Figure 2 shows a typical beta-gated gamma spectrum produced after a 3 hour wait period. The 30-keV peak contains signatures from ^{133}Xe, ^{135}Xe, ^{85m}Kr and ^{88}Kr, hence it is difficult to cleanly separate these isotopes using this region of the spectrum. After the two short-lived krypton isotopes have decayed, it is possible to separate out ^{133}Xe and ^{135}Xe as well as the two metastable xenon isotopes ^{131m}Xe and ^{133m}Xe.

A beta-gated gamma spectrum is shown in figure 2 that clearly shows ^{85m}Kr, ^{88}Kr, ^{133}Xe, and ^{135}Xe. These isotopes are the noble gas fission products generated from a 14-day neutron irradiation of the RXG. The short half-lived radiokryptons and ^{135}Xe have reached secular equilibrium after a couple of days of irradiation and the long irradiation time was chosen to maximize the ^{133}Xe yield.

Radioxenon From Neutron Irradiation

The production of neutron light radioxenons (those not produced via fission of TRU) may present unique challenges in identification of the standard four isotopes ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe if any of these additional isotopes are present in the sample. The additional radioxenon isotopes come from the neutron irradiation of natural xenon and the four of most concern are ^{122}Xe, ^{125}Xe, ^{127}Xe and ^{129m}Xe. Predicted activations and branching ratios indicate that these four isotopes may generate conversion electrons that may interfere with beta spectroscopy of the radioxenon isotopes from fission. Two of the isotopes are known to be produced for medical purposes. Xenon-127 and ^{122}Xe are routinely generated at the LANSCE facility in Los Alamos National Lab and the BLIP facility at Brookhaven National Lab in NY for medical uses.[1] For this reason the additional beta-gamma signatures will most likely be seen in some small number of xenon collection units located near the point of generation for these isotopes or where these isotopes are used. Table 1.1 below shows the major gamma and beta

Isotope	Half-life	Gamma-rays (keV)	Beta (keV)	X-rays (keV)	CE (keV)
^{122}Xe	20.1 hrs	148.6 (3.1%) 350.1 (7.8%)	IB 530 (<1.0%)	28-33 (78.6%)	5-24 (71%)
^{125}Xe	16.9 hrs	188.4 (54.9%) 243.4 (28.8%)	β+ 1467 (0.69%)	28-33 (100%)	5-80 (120%) 155 (6.4%)
^{127}Xe	36.4 days	172.1 (23.5%) 202.9 (68%) 375.0 (15.9%)	IB 457 (<1.0%)	28-33 (54.6%)	5-33 (69.3%) 90-125 (29.4%) 138-168 (84.2%)
^{129m}Xe	8.89 days	39.6 (7.5%) 196.6 (4.6%)		29-35 (126.5%)	5-40 (215%) 162 (63.3%) 191-197 (60%)

Table 1.1 Data taken from E. Browne, R.B. Firestone, V.S. Shirley (1986).

signatures that each of the four new radioxenon isotopes emit. The short lived isotopes and metastable isotopes have not been included as they have half-lives of 3 hours or less and so are very unlikely to have much strength after generation, emission, collection, processing and eventual counting in a beta-gamma cell.

Tests and Measurements

As part of an initial study into these isotopes as interference terms a limited irradiation of ambient stable xenon was conducted here at PNNL in 2000. A small flask of natural xenon was placed next to the intense (80g) PuBe and ^{252}Cf neutron sources used in the previous ^{235}U irradiated radioxenon generator experiment. After 4 days of irradiation the flask was removed, and approximately 5 ml of gas was injected into one of the ARSA counting cells. The β-cells were calibrated with the ^{137}Cs βγ technique and the NaI spectrometer was calibrated with a mixed multiline source, using γ-rays at 22, 59, and 88 keV.

Data was collected in beta/gamma coincidence mode and there were three distributions in the 2-D spectrum, corresponding to 30, 60, and 80 keV energy deposition in the NaI spectrometer (see figure 3A). The 60 keV group in particular had a good correlation between the energy in the β-cell and the NaI spectrometer. The distribution of the counts in the 30 keV region was more extended, but was definitely peaked and the 80 keV distribution was clustered primarily at low energies.

After about 4 days of counting, the cell was evacuated with a syringe and counted for 2-3 days. The count rate of the 30 keV distributions dropped by a factor of 10, comparable to the drop expected from the memory effect. While the drop in the 60 and 80 keV distributions was not as great, the signal to noise of these two groups is not as good. It is likely that the overall background in the cell contaminated these two groups.

Results and Analysis

Examination of the 2-d βγ spectrum after projecting the beta signal gated by appropriate γ energy cuts (figures 3B and 4) shows the corresponding beta plus conversion electron distributions. The 1-D gamma-gated beta spectrum analysis show that the 60 keV peak is due to a ^{125}Xe conversion, emitting two iodine X-rays (1 from the original electron capture decay and the other from the conversion process). This peak also bled into the 30 keV distribution, presumably from inefficiency of the NaI detector

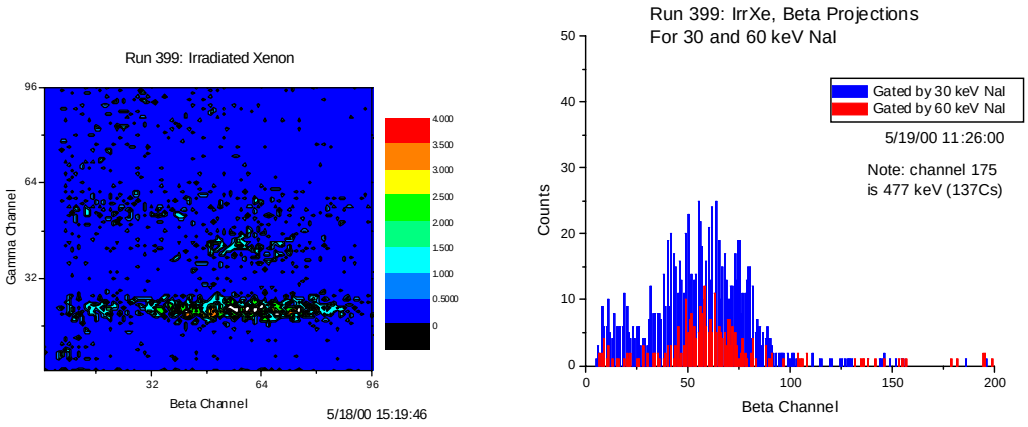

Figure 3A. This plot is the 2-d spectrum which clearly shows the three distributions in question. **Figure 3B**. The projected beta spectra from the 30 and 60 keV energy cuts. Note that the 60 keV spectrum does not extend to low pulse heights, and seems to be narrower than the 30 keV plot.

in detecting both the internal conversion x-ray and the conversion electron x-ray. Using the ^{137}Cs calibration technique for the beta energy axis, the energy of the β peak was ~160 keV (the predicted energy was 150 keV). There seem to be two other conversion lines, corresponding to fission product radioxenon. The data set is "sparse", however, and it is difficult to be quantitative. Comparing the 30 and 80 keV distributions it seems likely that the 80 keV gated spectrum has a contribution from ^{133}Xe. Most of the counts are at the lower end of the spectrum, as expected.

The activated xenon test will definitely spoil the resolution of the ARSA system for metastable analysis. There is another conversion line generated at 150 keV, filling in the beta spectrum. Additional tests would be convenient, however, to unambiguously identity this isotope as ^{125}Xe. While the energy spectrum is as predicted, the decay lifetime seems too long. Most of the decays occurred soon after irradiation, but there still seems an anomalously large contribution even after one week of decay. Considering that the limited number of counts, and no background subtraction, it is entirely possible that this apparently extended lifetime is a statistical anomaly.

Locally activated xenon offers a possible alternative to purchasing radioxenon from NIST or producing fission product samples at the Pullman TRIGA reactor. Considering activation of ^{125}Xe, the requisite one-week activating and counting periods are problematic. Activating a greater amount of this isotope would require that

activation would require either activation at Pullman or enriching the sample in ^{124}Xe. Natural xenon contains only 9×10^{-4} ^{124}Xe, and it is only observed due to its high neutron cross-section and short lifetime (17 hours). Enriching the sample to 25% ^{124}Xe would be 280 times more intense. Unfortunately, this isotope is expensive: 500 ml of 25% ^{124}Xe would cost $4600 after shipping and handling charges. A better choice might be a sample enriched in ^{132}Xe. This would be cheaper (^{124}Xe makes up 27% of natural xenon) and it would give both low energy β particles and a conversion line at 199 keV. This might be a useful spike sample for complete ARSA system tests, compared to detector calibration alone.

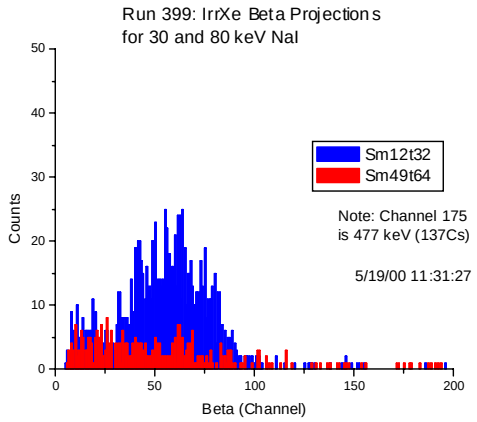

Figure 4. This plot is the comparison of beta spectra from the 30 and 80 keV NaI energy cuts. Note that both spectra extend to low pulse heights, consistent with ^{133}Xe. The broad peak in the 30 keV spectrum illustrates the difficulties of that irradiated xenon may present.

Conclusions

The production of the fission-produced radioxenons has been successful and a reliable supply of the short-lived ^{135}Xe has been demonstrated. Optimization of the process would be the next logical step and it is foreseen using other forms of uranium along with the potential to use plutonium would significantly enhance this process. For the neutron light radio xenon isotopes it has also been demonstrated that it is possible using existing equipment to produce two of the four light radioxenons . It is further noted that these isotopes represent additional signatures that may end up in radioxenon collection unit spectra that is near the point of use or generation. As such it is important to include the beta-gamma signatures as possible contaminants when extra peaks are detected in the beta-gated gamma spectrum. Since these isotopes have not been seen during the Freiburg tests of Phase II nor during the currently running Phase III tests it is not feasible to actually carry out detailed experimental examinations of the beta-gamma

coincidence spectrum at this time. It is however important for IMS analysts and others who will be responsible for the analysis of the radioxenon spectrum from all of the deployed systems to know of the potential signatures that these isotopes may have in some xenon collection units and not confuse the signatures with the more important fission produced radioxenon isotopes.

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[1] **Isotopes for Medicine and the Life Sciences, editors** S. J. Adelstein and F. J. Manning, National Academy Press, Washington D.C., p 62-65 (1995).