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Fluid Transfer Concentration of Airborne Radioxenon to Enhance Monitoring Capabilities

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Abstract

To facilitate airborne radioxenon monitoring, a xenon concentration method with potential advantages over current technology in simplicity, size, and cost has been developed. The concentration technique is based on the preferential absorption of heavy noble gases (krypton, xenon, and radon) by certain organic fluids. To implement this concentration technique, a radioxenon monitoring system requires three integrated sub-systems: 1) an absorption sub-system; 2) a degassing sub-system; and 3) a radiation detection sub-system. This study is focused on the characterization and optimization of the first two sub-systems. Measurements using a small prototype absorption tower have indicated a xenon removal factor of approximately 50% and the specific concentration at saturation of certain organic fluids to be about 2.5 times the specific concentration in the sampled air. Various techniques for degassing have been investigated, including heating, purging, agitation and vacuum. Ultrasonic agitation of a thin film in a strong vacuum has been shown to be an effective means of degassing the transfer fluid continuously. Various schemes for integrating all of the sub-systems are considered. Combining the small prototype absorption and degassing sub-systems should result in a transfer efficiency of about 33% and a single stage concentration factor of about 6.7.

1. Introduction

Atmospheric radioxenon is a primary indicator of the presence of a fission process. The type of source, typically an operating reactor or a nuclear weapons test, and the distance from the source to the detection system can be determined by measuring the ratios of radioxenon isotopes. However, detecting ambient radioxenon is complicated by relatively short half-lives and low atmospheric concentrations. The four isotopes of interest, ^{131m}Xe , ^{133}Xe , ^{133m}Xe , and ^{135}Xe , have half-lives ranging from 9.1 hours to 11.9 days. With limited time to accurately and

precisely detect these isotopes, an ambient xenon detection system should be able to perform measurements directly in the field. In addition, the system must be quite sensitive to the low concentrations present in the atmosphere, with specific activities on the order of tens of mBq per cubic meter. One method to enhance the sensitivity of a detection system is the extraction and concentration of radioxenon from large atmospheric samples. Relating the specific activities of these samples back to the volume of processed air would indicate actual concentration. Current technology to concentrate radioxenon primarily consists of cryogenic charcoal absorption [1]. This study develops an alternative means of concentrating ambient radioxenon based on a fluid transfer technique.

Fluid transfer concentration uses the affinity that certain organic fluids have for heavy noble gases (krypton, xenon and radon) to preferentially absorb these gases in the fluid. Subsequent degassing releases the absorbed heavy noble gases without the dilution of less soluble atmospheric gases. This concentration system consists of two sub-systems: 1) the absorption sub-system, and 2) the degassing sub-system. The overall radioxenon monitoring system consists of these two sub-systems integrated with a radiation detection sub-system, such that concentration and quantification are optimized. In this study, characterization and optimization of the concentration sub-systems will be addressed. Previous studies have evaluated the absorption properties of various organic fluids [2,3]. Corn oil was used with the sub-system prototypes in this study because of its relatively good performance, environmentally benign characteristics, and low cost.

2. Absorption

A cross-current absorption tower, as shown in Figure 1, was used to characterize heavy noble gas absorption by the organic fluid. A similar absorption tower design was used in preliminary characterization experiments [2]. The absorption tower used in this study was limited to about 50 cm in height because of fume hood restrictions, with a packed bed length of approximately 35 cm. Fluid was pumped into the top of the tower and then flowed down through the packed bed into the fluid reservoir. Air was pumped into a diffuser in the fluid reservoir and then flowed up through the packed bed. The packed bed consisted of 0.635 cm×0.635 cm cut polyethylene tubing.

Insoluble gases were released via the air outlet in the upper plenum. This absorption tower was used to measure the xenon removal factor of the absorption tower and the xenon concentration factor of the fluid.

The xenon removal factor is a measure of the fraction of xenon removed from a spiked air sample after passing through the absorption tower one time. To characterize removal factor, the once-through absorption sub-system, shown in Figure 2, was used. Various fluid and air flow rates were tested. Results indicate little dependence on fluid flow rate, but a stronger dependence on air flow rate and input xenon concentration. The best removal factor was approximately 50% with fluid flow of 0.25 lpm, air flow of 0.2 lpm and input ^{133}Xe concentration of 6.29 MBq/m³. The removal factor improves with lower air flow rates, lower input radioxenon concentrations and a greater length of packed bed.

The xenon concentration factor has been defined as the ratio of specific activity from absorbed radioxenon in saturated fluid to the specific radioxenon activity in the surrounding air. The concentration factor is not dependent on flow rates because it is determined at equilibrium saturation conditions. It also seems to be fairly independent of the input radioxenon concentration. For corn oil, the measured concentration factor was about 2.4 for 54.39 MBq/m³ of ^{133}Xe in surrounding air and about 2.5 for 36.11 MBq/m³ of ^{133}Xe in surrounding air.

3. Degassing

Removing the absorbed gas from the host fluid can be accomplished with a variety of techniques. Each method was evaluated for energy and time requirements and dilution of the degas product. Although the absorption mechanism is not fully understood, experiments seem to indicate that in addition to overcoming the standard desorption process dictated by partial pressures per Henry's law, a capture mechanism must also be overcome to cause degassing. Some sort of agitation, thermal or mechanical, could be used to release weakly trapped heavy noble gasses. Enhancing diffusional mass transfer out of the fluid could be accomplished through entrainment with a purge gas or by using a vacuum. Consequently, methods that have been investigated include heating, bubbling with a purge gas, ultrasonic agitation, vacuum and combinations thereof. These degassing techniques were

characterized using batches of fluid which had previously been passed through the absorption tower and thus contained absorbed radioxenon.

Experimental results where only heating was used to degas the fluid indicate that a complete degas would require about one hour with an approximately constant degas rate once the fluid temperature was above about 60°C. Consequently, the time and energy requirements were considered excessive for a continuously operating system, although heating would not dilute the degas product. While heating may effectively overcome the capture mechanism, it did not sufficiently drive desorption.

While bubbling with a purge gas in an open loop system was observed to be a relatively efficient degassing technique, the resulting dilution renders this technique ineffective for concentration purposes. Bubbling a purge gas through the fluid in a closed loop system was relatively ineffective, probably because of reabsorption in the fluid. Nonetheless, these air sparging results indicate that the mechanical agitation of bubbling combined with desorption-driving entrainment could be effective if replaced with less diluting methods.

To confirm that ultrasonic agitation would degas the fluid, a sonic disrupter was submerged about 3 cm deep in a beaker with 100 ml of fluid. Activity was monitored using a 5 cm×5 cm NaI(Tl) detector and temperature was monitored to verify any thermal effects. Results indicate that sonic agitation by itself is slow, with a complete degas requiring over 30 minutes for the 100 ml, at which point heating also became a factor. Nonetheless, cavitation and agitation were very evident, suggesting that ultrasonic agitation might be effective when combined with a technique to enhance desorption.

Vacuum was tested with 300 ml of fluid in a vacuum chamber. Activity was monitored using a 5 cm×5 cm NaI(Tl) detector. Results indicate very little degas with the 72 cm Hg vacuum alone. While the vacuum was creating a strong pressure gradient to drive desorption, there was no accompanying agitation. A magnetic stirrer was added to provide the agitation. A complete degas was realized in about 20 minutes after the magnetic stirrer was started and created some cavitating agitation. Further tests demonstrated that a thinner layer of fluid in the bottom of the vacuum chamber also increased degassing efficiency. This suggested that creating a thin film of the fluid, perhaps with a packed bed, would also enhance degassing. Aside from leakage, the vacuum technique does not dilute the degas product.

Using the results of the previous batch experiments, a prototype degassing sub-system was constructed for continuous flow experiments. This prototype combines a strong vacuum with a sonic disrupter housed in a flow cell that drains into an 20 cm packed bed, thin film tower. This vacuum chamber design, shown in Figure 3, was tested with 200 ml of fluid at 0.1 liters per minute continuous flow. After passing through the degassing chamber once, the sample was degassed $66.7 \pm 3.2\%$ with little heating or dilution. To optimize the degassing sub-system, a longer packed bed and multiple stages could be incorporated. Increasing the diameter of the chamber from the 5 cm diameter prototype would also minimize flooding that limits the flow rate to about 0.1 lpm.

4. System Integration

Given an optimized absorption tower and degassing chamber, these sub-systems must be integrated and optimized in a way that most effectively concentrates xenon for subsequent detection. Three potential concentration schemes are single pass, holdup and recirculation. All of these schemes consist of a continuous loop of absorption fluid being pumped between the absorption tower and the degassing chamber while air is pumped through the absorption tower. The schemes differ in the treatment of the degas product drawn off by the vacuum pump. The single pass system simply passes the degas product through the detection sub-system prior to venting. The holdup system retains the degas product in a chamber placed in front of the detector, with pressure increasing in this chamber over time. The recirculation system passes the degas product through the detection sub-system prior to combining it with air entering the absorption tower. The holdup system has the advantage of retaining the most degas product in the detection sub-system for the greatest amount of time. However, since any source chamber and vacuum pump have a limited pressure capacity, excess degas product must be vented. Venting excess degas product to the atmosphere, as in the single pass system, results in the system concentration factor of:

$$\frac{C_{deg}}{C_{air}} = \epsilon_{area} \frac{R_{air}}{R_{deg}} \quad (1)$$

where C_{deg} is the specific radioxenon concentration in the degas product, ml^{-1} ,

C_{air} is the specific radioxenon concentration in the input air, ml^{-1} ,

R_{air} is the volumetric flow rate of input air, ml/s,

R_{deg} is the volumetric flow rate of degas product, ml/s, and

ϵ_{tran} is the transfer efficiency: input Xe atoms per unit time/degassed Xe atoms per unit time.

However, if the excess degas product is recirculated back into the concentration system via a pressure relief valve as shown in Figure 4, the system concentration factor is improved. The differential equation defining the dynamic concentration of the recirculation system is given by:

$$V_{deg} \frac{dC_{deg}(t)}{dt} = \epsilon_{tran} (R_{deg} C_{deg}(t) + R_{air} C_{air}(t)) - R_{deg} C_{deg}(t) \quad (2)$$

where V_{deg} is the total volume occupied by the degas product (ml).

The general solution for this differential equation is given by:

$$C_{deg}(t) = \frac{\epsilon_{tran} R_{air}}{R_{deg} e^{\frac{(1-\epsilon_{tran}) R_{deg} t}{V_{deg}}}} \int e^{\frac{(1-\epsilon_{tran}) R_{deg} t}{V_{deg}}} C_{air}(t) dt \quad (3)$$

The system concentration factor for the recirculation system in steady state is given by:

$$\frac{C_{deg}}{C_{air}} = \frac{\epsilon_{tran}}{1 - \epsilon_{tran}} \frac{R_{air}}{R_{deg}} \quad (4)$$

This steady state solution shows an improvement factor over the single pass system of $1/(1-\epsilon_{tran})$. Of course, n multiple stages of concentration systems would yield a concentration factor of $(C_{deg}/C_{air})^n$, limited only by the number of stages. However, to minimize the number of stages required, each stage must be optimized.

5. Conclusions

The absorption tower characterized in this study absorbs about half of the radioxenon contained in input air. A fieldable absorption tower, not limited to 50 cm in height or to a single tower, should absorb a greater percentage

percentage (>90%) with a greater air flow rate [4]. The degassing sub-system characterized in this study demonstrated the ability to degas about 67% of the absorbed xenon continuously. A fieldable degassing unit consisting of several stages should also be able to improve on the percentage of xenon degassed. However, given only the tested laboratory scale units, approximately one third of the xenon contained in the input air will be transferred into the detection sub-system by the fluid. With measured input and degassing air flow rates of 4.0 lpm and 0.03 lpm, respectively, and a 33% transfer efficiency, the minimum system concentration factor for an unpressurized, recirculating source cell is about 6.7 for a single stage.

Fluid transfer concentration of radioxenon has potential advantages in simplicity, size and cost relative to current cryogenic distillation systems. Since the fluid transfer concentration system will also concentrate radon, the integrated system must be able to distinguish radon. Radon discrimination can be accomplished using pulse processing techniques [5], or possibly by fractionation of the xenon and radon in the degassing sub-system of the concentration system. Further study will include more optimization of the prototype concentration sub-systems, empirical characterization of the integrated concentration system, radon discrimination, and monitoring system calibration.

Acknowledgements

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References

- [1] P. Reeder and T. Bowyer, "High Sensitivity Detection of ^{133}Xe via Delayed Coincidence Counting," Pacific Northwest National Laboratories, Richland, Washington, 1997.
- [2] K. V. Liu, J. D. Gabor, K. C. Gross, and R. E. Holtz, "Fluid-Based Radon Mitigation Technology Development for Decontamination and Decommissioning," proceedings of the fourth international Conference on Nuclear Engineering, New Orleans, Louisiana, 10-14 March 1996.

- [3] M. Steinberg and B. Manowitz, "An Absorption Process for Recovery of Fission Product Noble Gases." *BNL 493 (T-115)*, Feb. 1958.
- [4] K.V. Liu, J.D. Gabor, R.E. Holtz, and K.C. Gross, "Fluid-Based Radon Mitigation Technology Development for Industrial Applications," *Proc. 1996 Chinese American Academic and Professional Conference.*, Ottawa, Canada (June 29 - July 2, 1996).
- [5] W. R. Russ, D. O. Stuenkel, J. D. Valentine, and K. C. Gross, "An Evaluation of Radioxenon Detection Techniques for use with a Fluid-Based Concentration System" submitted for presentation at the 1998 IEEE Nuclear Science Symposium, Toronto, Canada, November 1998

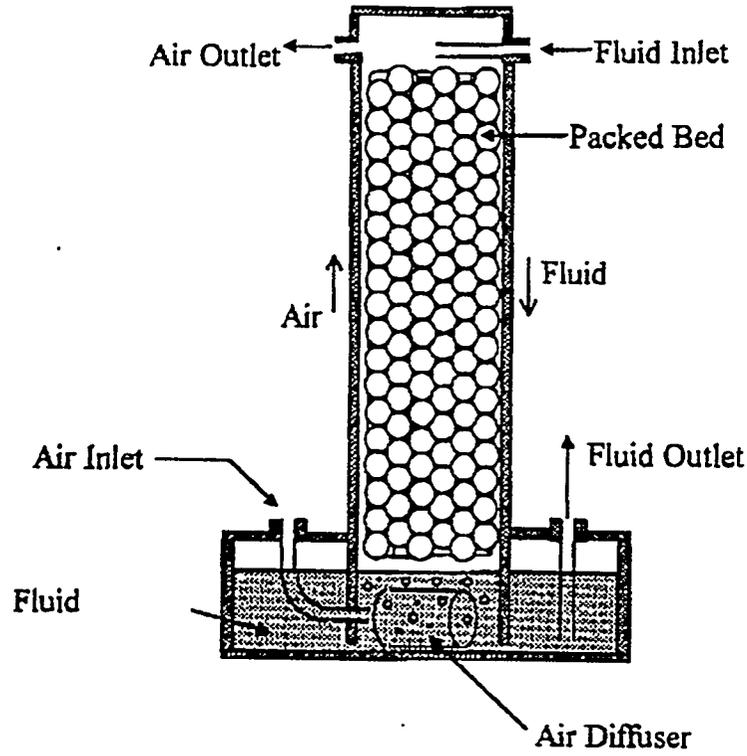


Figure 1 - Schematic of an absorption tower used to transfer xenon from the air to an organic fluid.

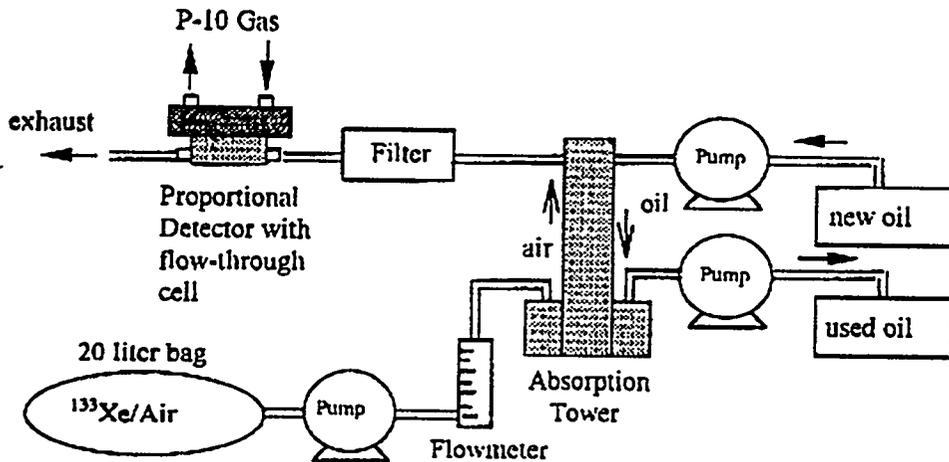


Figure 2 - Experimental configuration for measuring xenon removal factor of the prototype absorption tower.

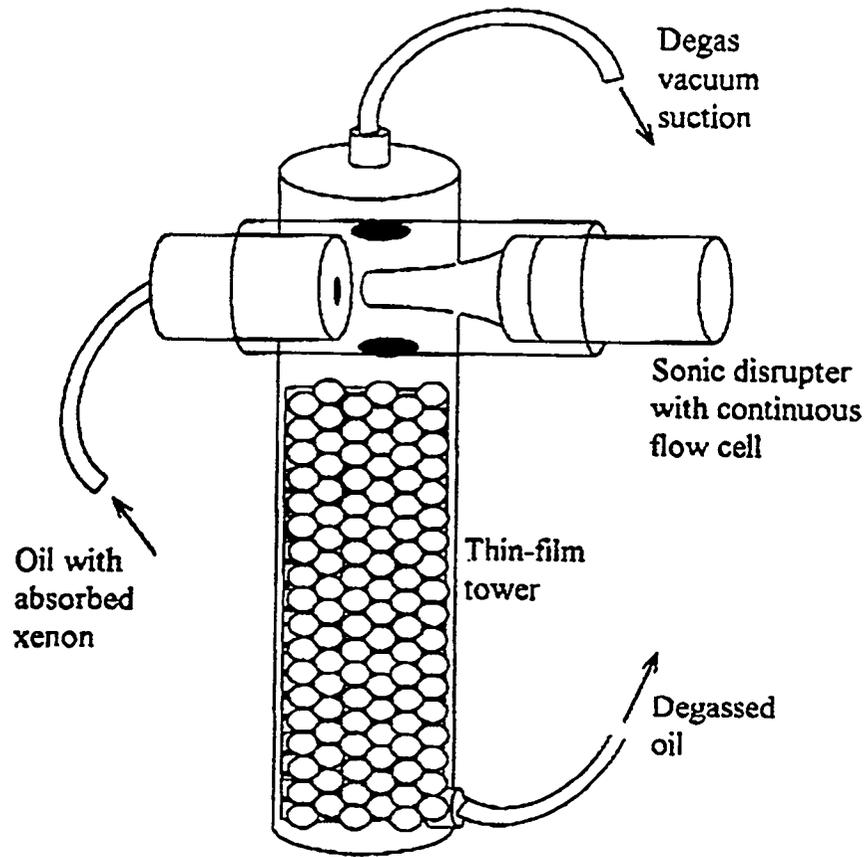


Figure 3 – Diagram of the degassing chamber constructed as a combination of sonic agitation with strong vacuum and thin-film tower with a strong vacuum.

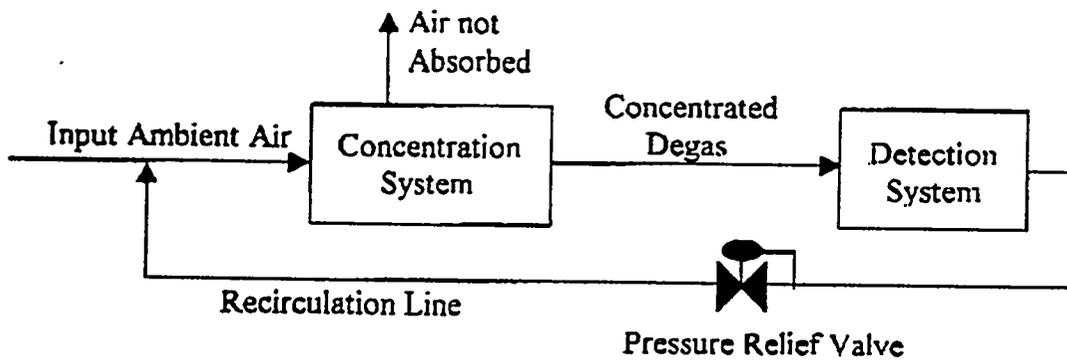


Figure 4 – Simplified schematic of combined holdup and recirculation scheme for system integration.