RADON REDUCTION EXPERIENCE AT A FORMER URANIUM PROCESSING FACILITY

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ABSTRACT

Approximately 6,200 cubic meters of waste containing about 2.0E8 MBq of radium-226 are stored in two large silos at the Fernald Site in southwest Ohio. The material is scheduled for retrieval, packaging, off site shipment and disposal by burial. Air in the silos above the stored material contained radon-222 at a concentration of 7.4 E5 Bq/L. Short-lived daughters formed by decay in these headspaces generated dose rates at contact with the top of the silos up to 1.05 mSv/hr and therefore complicate the process of retrieval.

A Radon Control System (RCS) employing carbon adsorption beds has been designed under contract with the Fluor Fernald to remove most of the radon in the headspaces and maintain lower concentrations during periods when work on or above the domes is needed. Removing the radon also removes the short-lived daughters and reduces the dose rate near the domes to 20 to 30 μSv/hr. Failing to remove the radon would be costly, in the exposure of personnel needed to work extended periods at these moderate dose rates, or in dollars for the application of remote retrieval techniques. In addition, the RCS minimizes the potential for environmental releases.

This paper describes the RCS, its mode of operation, and early experiences. The results of the test described herein and the experience gained from operation of the RCS during its first phase of continuous operation, will be used to determine the best air flow and air flow distribution, the most desirable number and sequence of adsorption beds to be used and the optimum application of air recycle within the RCS.

INTRODUCTION

One element of the Fernald Closure project is the removal of K-65 material from two large silos on site. The K-65 material is the residue from the extraction of uranium from high-grade ore. The chief radiological material remaining is radium-226 at a concentration of about 1.5 E4 Bq/g. In total there are about 9,000 metric tons of K-65 material, containing 1.74 E8 MBq of radium.

The silos are concrete structures 24.4 m in diameter by 8.23 m high on the sides. The height in the center of the dome is 11.0 m. A soil berm is piled up against the sides of the silos, providing shielding and structural stability. The silos had a steel-reinforced plywood cap to redistribute the load on the dome, and a foamed cover to retard the release of radon

Inside of the silos radon generated by the decay of radium in the K-65 material migrates to the headspace and decays there. Two of the short lived radon daughters, Pb-214 and Bi-214 emit gamma radiation. Some of the radon escapes from the silos, by diffusion through the concrete, or via micro-cracks with changes in the weather. In addition, the gamma radiation from the daughters makes both of the domes large sources having moderate dose rates.

One effort at mitigation was implemented in 1987. A radon treatment system (RTS) was constructed in which the headspace air was drawn from the silo headspaces, passed through a carbon bed and returned to the silos. This system was operated when access to the silo domes was required for sampling or maintenance. It was considered to be a temporary control system.

A two-foot layer of bentonite grout was added to the silos in 1991 to provide a more permanent solution. This addition did retard the migration of radon into the headspace, reducing both leakage and dose rate. However, during recent years the bentonite has dried and cracked and the radiological conditions have returned to near their previous highs. The concentration of radon in the headspaces reached 7.4 E5 Bq/L and the contact dose rate on the silos was as high as 1.05 mSv/hr. As a matter of perspective, 7.4E5 Bq/L is 670,000 times the Derived Air Concentration (DAC) for radon with daughters present.

Because of the high concentrations of radon, the moderate dose rate, and the large volume of K-65 material, dealing with the radon is prerequisite to the removal, treatment, and packaging of the K-65 material.

APPROACH

The success of the temporary RTS formed the basis for a decision to build a larger radon control system effective enough to manage the radon for the entire project. At first the RCS operation will need to remove radon from the silo headspaces to facilitate work in the area. In phase 2 the RCS will control radon released while the K-65 material is slurried from the silos to interim storage tanks in the Transfer Tank Area (TTA). The RCS will also be needed to manage the radon in the process off-gas during the third phase when the material is slurried out of these tanks and treated.

The principle behind the operation of the next-generation radon control system is based on slowing the radon down as it passes through a bed of activated carbon (since radon is not irreversibly adsorbed) for long enough for most of it to decay. The effect of the bed on the concentration of radon described for the steady state by using the following equation [1]. Variables are the mass of carbon, the ventilation air flow rate, and the dynamic adsorption coefficient for the carbon.

$$C_{out} = C_{in}(\exp(-\lambda KM/R)) \quad \text{(Eq. 1)}$$

C = Radon concentration entering the bed (in) or leaving the bed (out) (Bq/L)
λ = Decay constant for radon (min^{-1})
K = Dynamic adsorption coefficient (L/g)
M = Mass of carbon (g)
R = Flow rate in carbon bed (L/min)
$1-(C_{out}/ C_{in})$= Fraction radon removed

Effective performance in reducing radon concentrations, i.e., C_{out}/C_{in}, over a long period of time is achieved by using a large mass of carbon, by minimizing the flow of air through the bed, and by achieving a high dynamic adsorption coefficient. Studies have shown that the dynamic adsorption coefficient increases with decreases in both temperature and humidity. For example, the dynamic adsorption coefficient is reported to increase from 6.1 L/g to 13.6 L/g when the temperature is decreased from 24°C to 5°C [1]. Similarly, the dynamic adsorption coefficient reportedly decreases by 50% with an increase in relative humidity from 15 to 30 % [2]. A 50% decrease in the dynamic adsorption coefficient corresponds in a 7-fold decrease in the ability to hold up radon.

The continuity equation is applied to assess the effect of drawing air from a silo, passing it through a bed and returning it to the silo in the following equation:

$$dC(t)/dt = E/V - \lambda C(t) - Q\eta C(t)/V \quad \text{(Eq. 2)}$$

C(t) = Headspace concentration (Bq/L)
E = Transfer rate from K-65 solids into the headspace (Bq/min)
λ = Decay constant for radon (1.26 E-4 min^{-1})
Q = Flow rate through headspace (L/min)
V = Volume of headspace (L)
η = Fraction of radon removed by carbon $(1-(C_{out}/ C_{in}))$

Equation 3, the solution shows the effect of the recycle flow in the headspace and the effectiveness of the carbon beds on the radon concentration there.

$$C(t) = (E/V\alpha)(1-\exp(-\alpha t)) + C_o(\exp(-\alpha t)), \quad \text{where } \alpha = (\lambda + Q\eta/V) \quad \text{(Eq. 3)}$$

A review of this equation shows what to expect in the headspace at key times. At first, before efforts to mitigate the radon, later, when the RCS is turned on, and much later after the system has been on long enough to reach steady state again. At the first key time $t = \infty$, and Q = 0. The

concentration in the headspace then is an equilibrium between the rate that radon is transferred into the headspace from the body of K-65 material (E) and the radioactive decay rate of radon in the headspace (λ). Leakage from the headspace is relatively small and considered insignificant. At the second key time when the RCS is first started, the reference time is restarted (t = 0) and Q is no longer zero. The initial concentration of radon, $C_{o,}$ decays away exponentially as the negative exponent is increased by Qη/V. New radon enters the headspace (E) and is returned during recycle, Qη. However, the additions are too small to mitigate the decrease of the initial radon present. Finally steady state is reestablished. The initial radon becomes insignificant, and the headspace concentration is at an equilibrium between the radon entering E and the radon leaving, VC(t)(λ + Qη/V).

With these principles in mind the design requirements for the system were developed to do the following:

-maintain a negative pressure in the headspaces of the silos and other process vessels
-limit radiation fields adjacent to the silos and the RCS itself
-prevent uncontrolled releases of radon
-provide for process control via both local and remote monitoring
-provide for isokinetic sampling and monitoring of the stack discharge
-maintain off-site concentrations of radon below and annual average of 0.5 pCi/L

The concept for the initial operation of the RCS (Phase 1) is shown in Fig. 1.

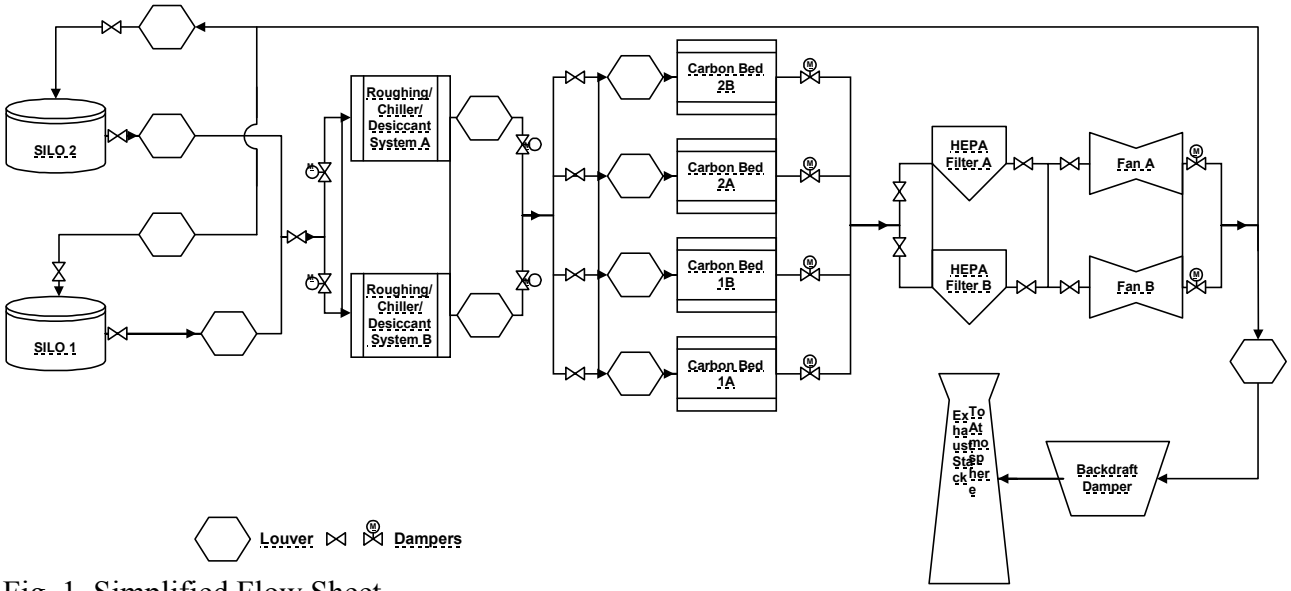

Fig. 1. Simplified Flow Sheet

One of the fans pulls air from the silos through the desiccant dryers and the carbon beds. Most of the radon decays in the beds and is removed from the air streams. The treated air is returned to the silos or exhausted via the stack. Louvers and dampers control the flows and HEPAs remove particulates (if any) before discharge.

FACILITY DESCRIPTION

The construction of the RCS began in June, 2000. The features of the system include fans, two roughing filters, two desiccant dryers, four carbon beds, two HEPA filters, one stack monitor, and ductwork as necessary to direct the flow of air. The fans provide the motive force by pulling the air from the silos to the RCS treatment equipment located in the RCS building. Here the inlet duct is branched so that flow can be diverted into either of two roughing filters on the first floor of the RCS building, as shown in Fig. 2 the simplified plan view of the first floor.

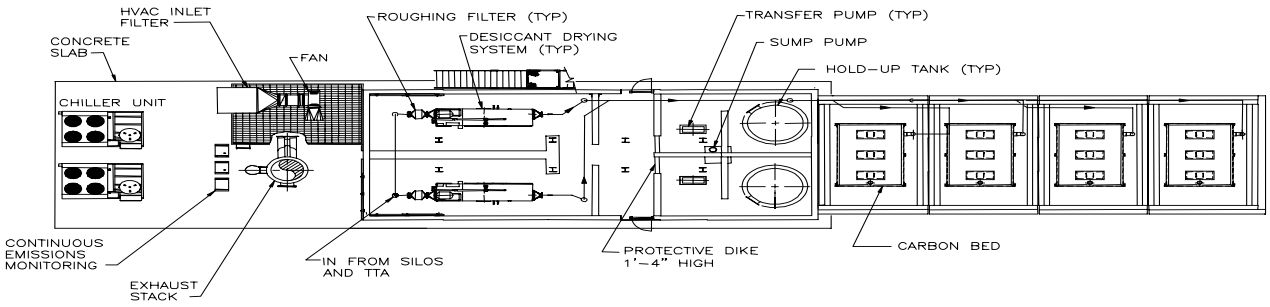

Fig. 2. First Floor Plan View of RCS

The roughing filters remove a nominal 95 percent of the particulate material carried in the air stream, including most of the radon daughters, keeping them out of the rest of the system. The buildup of radon daughters on the filter generates substantial gamma radiation, and the vault housing the filters and the desiccant dryers is shielded and protected as a high radiation area.

From the filter the air immediately flows to a desiccant dryer where it is cooled, dried, and cooled again. The first cooling step reduces the air temperature to 7^{o}C and condenses out most of the moisture. The condensate is pumped to one of the 1.14 E4 L Condensate Storage Tanks for batch transfer to other water treatment facilities.

Drying is accomplished as the air passes through a desiccant wheel containing fluted passages that are impregnated with silica gel. Heat generated during the adsorption of water is eliminated and dissipated in a second cooling step. The air leaves the desiccant dryer at about 5^{o}C and 15% relative humidity (0.8 grams of water per kilogram of bone dry air). The chilled water compressor and heat exchanger associated with the coils in the desiccant dryer reside on a pad outside the west end of the building. Each of the desiccant dryers is rated at 2.83 E4 L/min and has the capacity to continuously recharge the desiccant wheel by driving the moisture off for recycle within the system.

The air stream flows from the desiccant dryer directly into the carbon beds. The beds are 3 x 3 x 4.5 m and contain approximately 18.2 metric tons of activated carbon each. The beds are operated in parallel and any combination of the beds may be selected for use at one time. The

design flow for the beds is 1.42 E4 L/min each. The beds are divided into three sections each with an overall area for flow of 1.5 m x 3 m. Air flows up through the first and third sections and down through the second. The activated carbon was specifically selected for its ability to adsorb radon to more effectively slow the passage of the radon through the beds [3]. The air exiting the beds at first is depleted in radon. Later, by the time the radon has traversed the bed, much of it has undergone radioactive decay and the original concentration is reduced by that factor (Radon-222 has a half-life of 3.82 days). While the radon is slowly traversing the beds and decaying equilibrium activities of its short-lived daughters, Po-218, Pb-214, Bi-214, and Po-214, build up. Gamma radiation from the Pb-214 and Bi-214 is sufficient to generate dose rates in excess of 1 rem/hr. Therefore, each bed is enclosed in a vault shielded with two feet of concrete. Because of the importance of keeping the carbon beds cool, the vaults are cooled using a dedicated air handling unit. The vault areas are not accessible to personnel.

By the time the air exits the beds the radon concentration has been reduced. This treated air then passes through one of two HEPA filters housed on the second floor and enters the fan housing. Figure 3 is a simplified plan view of the second floor.

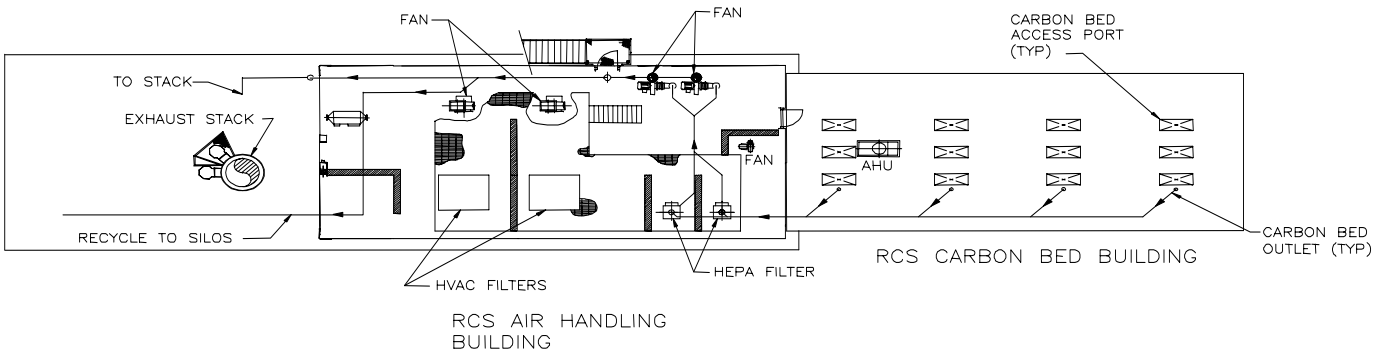

Fig. 3. Second Floor Plan View of RCS

The fan output is recycled back to the silos or discharged directly to the stack. Note that in future phases the air may also be recycled to tanks in the TTA. However, the design is for at least five percent of the fan output to flow to the stack.

The RCS building is served with an independent HVAC system that pulls fresh air into the building, sequentially through the various operating areas, through HEPA filters, and exhausts this ventilation air to the stack (along with the air discharged from the RCS).
The flow in the stack is composed of 2.83 E5 L/min from the building ventilation system and 1.0 E2 to 5.66 E4 L/min from the RCS. The stack is 45.7 m tall and equipped with an isokinetic monitor to assess the emissions of both particulates and radon.

HOT TESTS OF THE RCS

At the conclusion of construction and cold operability testing two significant questions remained unanswered. First, how uniform would the flow through the beds be, that is, would there be channeling, and if so, how much? If too much air were to pass through the beds via channels,

e.g., preferential paths, the radon would not be effectively adsorbed and bed performance would be limited. Secondly, what would the actual dynamic adsorption coefficient be? This factor had been based on a laboratory scale measured for the carbon selected, but would performance on an industrial scale be as good?

The first hot test was begun in December, 2002. An air flow of Eight thousand five hundred liters per minute per silo was chosen to conduct the test. This flow was well within the designed flow capacity of the carbon beds (1.42 E4 L/min per bed) and was enough to achieve the desired reduction in headspace concentration (according to Equation 3). The air was cooled to 5°C and dried to a relative humidity of 15%. Almost immediately the concentration of radon in the silo headspaces began to drop, and before the test was terminated 17 hours later the concentrations in the silo had dropped more than 95% from 7.4 E5 Bq/L to less than 3.7 E4 Bq/L. Although the system was designed to recycle as much as 100% of the purified air back to the silos, only eighty-five percent of the flow through the carbon beds was recycled, with the remainder exhausted to the stack. This minor reduction in recycle air flow made it easier to control the pressure in the silos. A controlled influx of ambient air made up for the loss. Figure 4 shows the change in the concentration of radon in the silos during the first hot test and the days following.

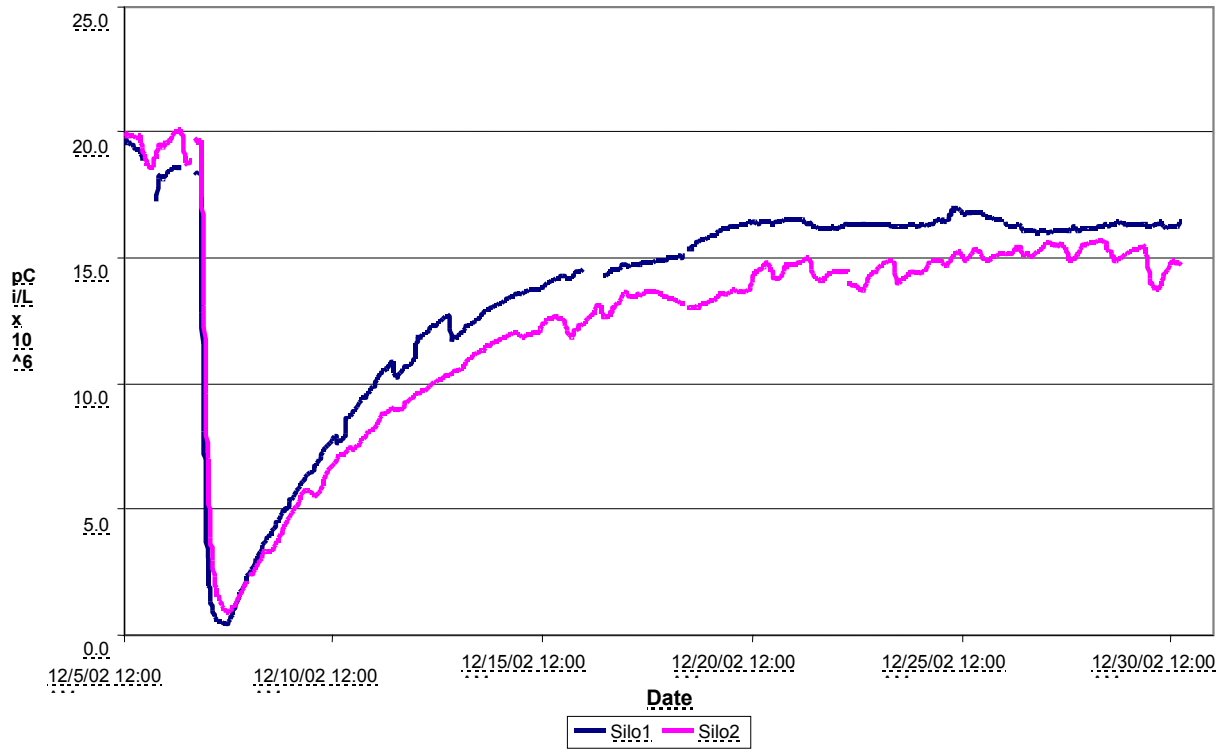

Fig. 4 Silo Headspace Radon Concentration in Hot Test #1

As usual, after the first time a system is run there are improvements to be made. Several months were taken to reduce excessive recycling of the desiccant dryers, to better manage the condensate formed in the hoses that bring air from the silos to the RCS, and to reroute the return lines from the radon monitors – which had reentered the system downstream of the carbon beds.

The second hot test was designed to answer the still unanswered questions about extended performance. It was begun in April 2003 with two beds and other initial conditions much as before. However, this time 1.42 E4 L/min (the maximum design flow per bed) was withdrawn from each silo to shorten the time necessary to reach steady state. The air was cooled to 5°C, dried to 15% RH and passed through two carbon beds. And again, 85 percent of the air exiting the beds was recycled to the silos. The radon concentrations in the silos, that had increased to near 7.4E5 Bq/L by April, were swiftly reduced as before. Then, to avoid the risk of an abrupt peak in the concentration of radon exiting the beds, the flow was switched to the other two beds.

The dynamic adsorption coefficient was expected to be about 13 L/g based on measurements made during the procurement of the carbon. Based on this, breakthrough of the radon was expected in about 2 weeks (Of course the exiting radon concentration would be less than the input concentration by a factor of 12.6 because of the decay during that time.). Consequently, it was of major concern when the first significant radon was seen exiting the beds after just four days. This is shown in Fig. 5.

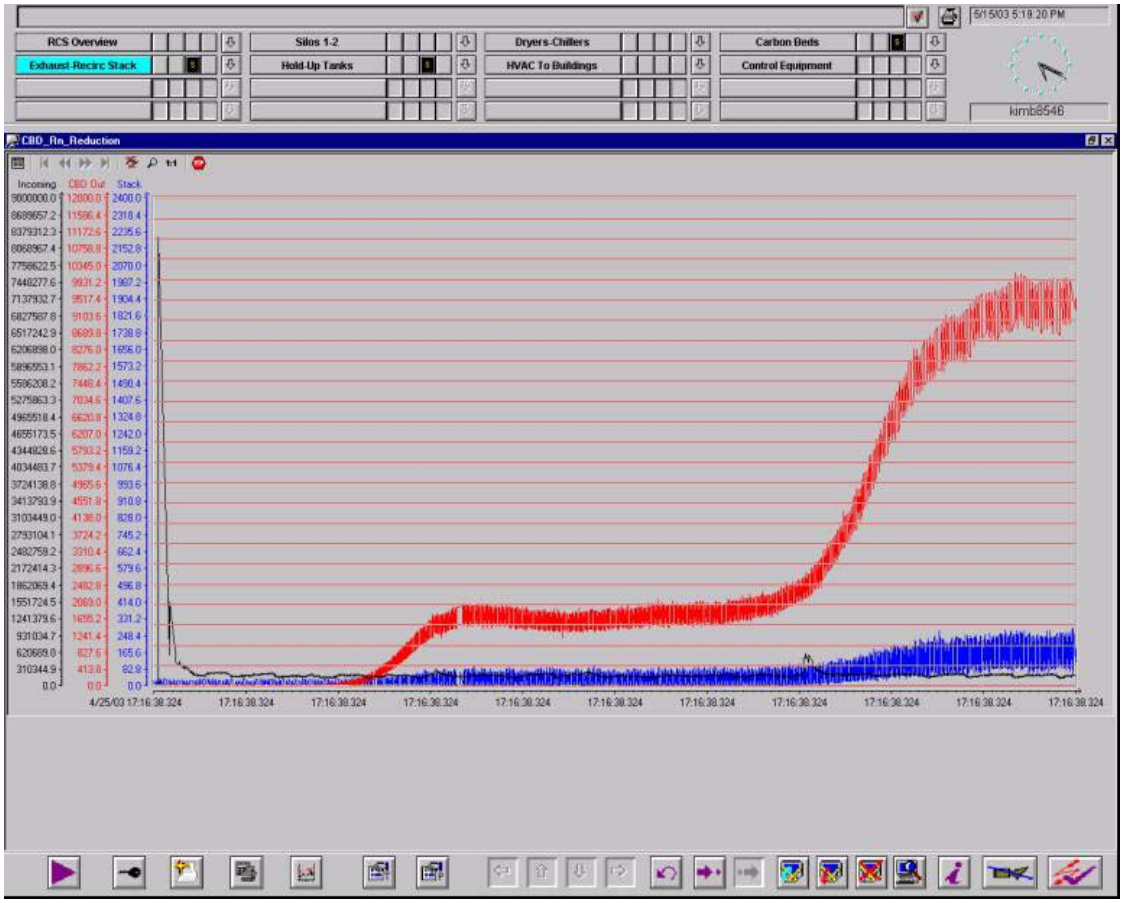

Fig. 5. Radon Concentrations in Hot Test #2

This figure is a reproduction of the Control Room computer screen showing the first 20 days of Hot Test #2. There are three traces shown each with its own scale. Barely visible on the left is the headspace concentration that drops from 7.4E5 Bq/L to about 1.5 E4 Bq/L and then stabilizes there for the duration. The second trace is the carbon bed output concentration, showing two plateaus at about 67 and 350 Bq/L. The concentration of radon in the stack is the third trace, following the carbon bed outlet concentration, but lower by a factor of about 50.

A review of Fig. 5 shows that the radon concentration initially increased to about 0.5 % of the stabilized input concentration of 1.5E4 Bq/L and then leveled off. However, this was not true breakthrough. At true breakthrough the ratio of the output to input concentration is given by the following:

$$C_{out}/C_{in} = \exp(-\lambda t) \qquad \text{(Eq. 4)}$$

where "t" is the time to breakthrough. True breakthrough was not the case for the first plateau where "t" is about 6.5 days (or 9,360 minutes), i.e., 67/1.5E4 does not nearly equal exp((-1.26E-4)(9360)). So the test continued without change. Finally, after 12 more days true breakthrough did occur. Here the ratio, i.e., 350/1.5E4 = 0.023, approached the value predicted by Equation 4, i.e., 0.035.

The first radon seen was attributed to channeling, where about 2 % of the air had found a preferential path and the radon it contained was poorly adsorbed as a result. The radon seen on true breakthrough was used to determine the actual (as opposed to theoretical) dynamic adsorption coefficient It was found to be about 22 L/g according to Equation 1, much better than the 13 L/g expected. The last element of Hot Test #2 was to repeat the sequence using the other two beds. The results were almost identical, e.g., channeling after four days and an actual dynamic adsorption coefficient found from breakthrough after about 19 days to be 20 L/g.

Hot Test #2 was successful in answering the two key performance questions. That is, channeling had little effect on the overall performance of the beds, and the system had performed beyond design expectations.

During Hot Test #2 the equipment (fans, desiccant dryers, flow controllers, e.g., dampers and louvers) was exercised. They responded well and the RCS was able to maintain the pressure in the silos at -12.7 mm H_2O as desired, and the desiccant dryer was able to condition the air to meet $<5^oC$ and <15% RH targets.

OPERATIONS

Because of the success of Hot Test #2 and schedule pressures, the RCS was transitioned directly from the test phase to operations. Physically, there was little difference between the end of Hot Test #2 and the beginning of operations. Readiness was demonstrated, flow was switched to the other set of beds, and the airflow rate was dropped to about 7.9E3 L/min.

The radiological effects of Hot Test #2 and early operations were impressive as the headspace radon concentration was reduced and a negative pressure was maintained in the silos. The

escape of radon through the domes that had been estimated recently to be as high as 1.9 MBq/yr was essentially eliminated. Ambient radon monitors in the area near the silos saw a decrease in radon concentrations from typical historically recorded concentrations between 0.2 to 0.4 Bq/L to concentrations typically less than 0.03 Bq/L. In addition, the dose rates in contact with the dome surfaces were reduced from as high as 1.05 mSv/hr to 0.02 to 0.03 mSv/hr. This in effect removed two 470 m^2 extended sources of gamma radiation and dropped the area dose rates to near background levels of 0.008 μSv/hr. The reduction of the gamma radiation from the domes, along with the reduction in outdoor radon allowed administrative controls to be relaxed, saving cost and schedule as construction was active in this area. The controlled area was reduced from about 60,000 m^2 around the silos to an area enclosing only the berms and the silos themselves (only 6000 m^2).

Work on (and over) the domes progressed expeditiously under these improved conditions. The first activity was the removal of water that had collected under the caps, and then the caps themselves. Bridges were installed over the domes and holes were cut for the installation of risers. The risers were to accommodate the installation of the sluicers and the slurry pump necessary to slurry out the K-65.

A FORWARD LOOK

The start of transfer operations is imminent, and with it, new challenges are anticipated. First, at the present only about 1% of the radon is in the headspace. The other radon remains trapped, in the particles where it was produced, or at-large in the 11 m deep bed of K-65 and bentonite. When mining begins as much as 25 times more radon will be released to the headspace air. In addition, the RCS will have to service the receiving tanks in the TTA and each tank and silo will have three service modules connected to the risers. These modules will be subject to contamination as radon diffuses up from the silos or tanks and decays. Finally, the RCS will have to support the treatment and packaging of the K-65 material since the radon generated will have to be managed until the waste is solidified and sealed in its shipping container.

Operational margin is needed to meet these challenges. Some is available because only two beds have been used at one time. Using four beds will more than quadruple the capacity to control the radon. In addition, the flexibility of the system, allowing one to alter the airflow through any particular silo or tank provides margin. During times when only one silo is active, airflow through it can be increased to improve local conditions at the expense of other areas where there is no current work. Finally recycling some of the air is an option, but at the cost of greater contamination in the associated modules.
All-in-all, the performance of the RCS indicates that there is sufficient margin to meet the challenges, moving the K-65 material out of the silos into the new tanks, and treating and packaging it while still protecting the workers and the public.

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