

FLUID-BASED RADON MITIGATION TECHNOLOGY DEVELOPMENT FOR INDUSTRIAL APPLICATIONS

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ABSTRACT

The objective of the radon mitigation technology development effort is to develop an efficient and economical radon gas removal technology based on a fluid absorption process. The technology must be capable of cleaning up a wide range of radon gas stream concentrations to a level that meets EPA gas emission standards for residential and industrial applications.

Argonne has recently identified a phenomenon that offers the possibility of radon recovery from the atmosphere with high efficiency at room temperature, and radon release at slightly elevated temperatures (50–60°C). Such a device would offer numerous substantial advantages over conventional cryogenic charcoal systems for the removal of radon.

Controlled sources of radon in Argonne's radon research facility are being used to quantitatively assess the performance of a selected class of absorbing fluids over a range of radon concentrations. This paper will discuss the design of laboratory- and engineering-scale radon absorption units and present some preliminary experiential test results.

INTRODUCTION

Radon gas (Rn-222) is a major contributor to the natural radiation exposure of the average individual in the U.S. and is considered the most serious health threat of all the known indoor air contaminants. Past EPA studies [1993] have indicated that up to 14,000 deaths occurring annually in the U.S. may be attributed to indoor radon exposure. The removal of radon gas from nuclear facilities, uranium mines, and industrial sites has received increased attention.

An early goal of this work was to establish process feasibility, and then determine optimal performance parameters such as flooding characteristics, saturation capacities, Henry's Law coefficients,

fluid aging stability, packing material physical characteristics, absorption and desorption rates, etc., for a fluid-based system for removal and isolation of radon. The approach was to use controlled sources of radon in Argonne's radon research facility to quantitatively assess the above performance parameters for a selected class of absorbing fluids over a range of radon concentrations. Degassing methods considered were thermal (already proven in Argonne experiments), mechanical agitation, thin film flow, ultrasonic, and microwave.

The engineering development effort builds upon the promising laboratory equilibrium radon removal results obtained using an oil-based liquid absorbent. Initial absorber tests focused on confirming the proof-of-concept in a laboratory-scale packed absorption tower (3-in. diameter bed). Later tests will be conducted with 3-in. dia. absorption/desorption system and an engineering pilot scale unit (12-in. diameter bed) which will provide information for the commercial-scale radon removal systems.

This paper describes results obtained from static laboratory experiments, dynamic flow tests with a laboratory-scale apparatus, the design of engineering-scale, and commercial-scale radon removal units.

LABORATORY STATIC EXPERIMENTS

Static experiments were conducted for both absorption and desorption of radon in oil. The main emphasis during the initial study has been on the absorption of radon in oil. The results for degassification of radon in oil are preliminary. Although a variety of oils have been examined, most work has been conducted with Mazola® corn oil.

Concentration Factor Measurement

The concentration factor, defined as the radon concentration attained in oil divided by the radon

concentration of the input air stream, has been used as the first criterion to examine the radon absorption capability. During absorption testing, small volumes of oil (50–100 ml) were saturated with 17 l of air containing a known concentration of radon. The uptake of radon by the oil was then measured. Radon-free air was bubbled through the oil at a rate of 1 l/min. The air was passed through a dry ice-chilled charcoal trap to remove the radon. The radon from the charcoal was transferred to a Lucas cell scintillation chamber and counted. Mazola® corn oil and different vegetable and hydrocarbon-based oils were tested. Results from these tests are summarized in Fig. 1. It is clearly shown that 3-in-1 oil has the highest concentration factor (9.69) of all oils tested. Concentration factors between 8.1 and 8.69 were obtained in the Mazola corn oils tests.

Degassing Test Results

Earlier tests have shown that all radon can be removed from oil using a thermal degassification method with air purging. Degassing tests were also conducted on ultrasonic radon removal. The ultrasonic degassing results were assessed in terms of a radon removal factor defined as the radon removed divided by radon initially present. For sonication alone, the best removal factor achieved was 0.45. With air purging, the removal factor increased to 0.6. The highest radon removal factor, 0.77, was obtained when the ultrasonic degassing procedure was conducted in conjunction with air purging and partial vacuum conditions.

LABORATORY-SCALE EXPERIMENTS

A laboratory-scale absorption tower was constructed to obtain data on mass transfer rates and flow characteristics. The tower, which was constructed of Plexiglas®, has a 3-in. I.D. and either a short 16-in. high or tall 42-in. high packed-bed test section plus flow meters, a differential pressure gauge, and air/oil pumps. The absorbing liquid (corn oil in the initial tests) was circulated by a peristaltic pump. Raschig rings with 1/4-in. and 3/8-in. diameter were used for tower packing.

Measurements of radon removal were made with this apparatus in the Argonne Radon Research Facility. A summary of tests with the 1/4-in. Raschig rings is given in Table 1. It is clearly shown that the oil-based absorption column can remove the majority of radon gas in the air stream, up to 98.7%, with a 42-in. packing column under certain flow conditions.

The laboratory-scale radon absorption tower has been modified to include a radon desorber (Fig. 2). The desorber is made of a long 2-in. diameter copper

pipe with 1/4-in. diameter copper Raschig rings. The desorber is electrically heated with clamp heaters. A portion of the copper tubing leading to the desorber is also heated electrically. This arrangement will pre-heat the oil as it is flowing into the desorber. The radon will be absorbed in oil at room temperature in the absorption tower and be stripped in the desorber at a higher temperature as shown in Fig. 2. The oil is returned to the absorption tower after it is cooled down to the room temperature.

ENGINEERING-SCALE EXPERIMENTS

The design and construction of an engineering-scale absorption tower has been completed. This tower is 12-in. in diameter and 80-in. high. Testing of an engineering-scale unit is needed to advance toward the commercial-scale design required for industrial radon mitigation.

The engineering-scale absorption tower assembly is shown in Fig. 3. The tower consists of four 19.7-in. long glass sections. The packing will be supported on a Norton Model 809 support plate. Six different sizes of spray nozzles with flowing ranges from 0.74 to 76 gpm will also be tested. Four types of Norton metal packing No. 1 and No. 1-1/2 Hy-Pak, and No. 25 and No. 40 IMTP (1- to 1-1/2-in.) will be tested. The pressure drop measurement across the packing bed as a function of the air flow rate without oil present is shown in Fig. 4. The pressure drop increases from 0.2 in. water to 0.9 in. water at an air rate about 100 CFM when oil, ~23 gpm, is flowing down in the packed column.

This unit will be used for Radon 220 removal in the fan loft at Argonne National Laboratory, Building 200. Radon 220 is in the stack release from cell A-3. Because Radon 220 decays rapidly ($t_{1/2} = 55.6$ s) and will decay within the absorbent oil as it circulates through the system, no provision will be made to desorb (strip) the radon from the absorbent. During the course of testing, the exhaust air and oil flow rates will be varied and each of the packings will be tested. The theoretical radon removal efficiency from this absorption tower is presented in Fig. 5. The analytical results will be verified with the measurements in ANL—Building 200.

SCOPING STUDY OF A COMMERCIAL FACILITY

A parametric study based on the principles described above was conducted in order to scope the size and operating conditions to be expected in the full-scale design of a radon absorption tower based on the needs of the Department of Energy (DOE) Fernald facility. The computer program, ISGA by Sherwood, Pigford, and Wilke [1975], was modified

TABLE 1
RADON ABSORPTION IN A LABORATORY-SCALE TOWER

Conditions: 3-in. I. D. Plexiglas® tower
3.5 ft depth of 1/4-in. Raschig Rings
Inlet radon concentration in air: 289.25 pCi/ℓ
Inlet radon concentration in corn oil: 0 pCi/ℓ

Test No.	Rn in Air Out, pCi/ℓ	Air Flow, ℓ/min	Oil Flow, ℓ min	Transfer Unit Number	Transfer Unit Height * ft	Radon Removal, %
38	65.55	1.0	0.259	1.84	1.90	77.3
39	53.74	1.0	0.259	2.13	1.65	81.4
40	40.04	1.0	0.442	2.28	1.54	86.2
41	53.14	1.0	0.442	1.93	1.82	81.8
42	31.43	0.5	0.442	2.38	1.47	89.1
43	16.41	0.5	0.442	3.11	1.13	94.3
44	3.70	0.5	0.259	5.14	0.68	98.7
45	8.81	0.5	0.259	4.07	0.86	97.0
46	117.06	2.0	0.259	1.27	2.76	59.5
48	116.60	2.0	0.259	1.28	2.74	59.7

*Based on gas phase controlling.

and employed in this study. The off-gas flow was assumed to be 250 CFM at 88°F containing 38,550 pCi/ℓ radon in air. A radon decontamination of the gas stream to a level of 1100 pCi/ℓ (97% removal) as required for EPA compliance, was assumed in the calculation.

In these initial scoping calculations, the effect of the following parameters was assessed:

1. Packing size,
2. Transfer unit height,
3. Radon gas solubility, and
4. Flooding and absorption design calculational factors.

The tower was considered to be filled with Raschig rings. Packing size varied from 1/4 in. to 3 in. The height-of-transfer units for both gas phase and liquid phase transfer were varied over an arbitrary range of 0.25 to 1.75 from a reference value.

The tower diameters were established by the flooding rates dependent on the packing size. The tower diameters have a range from 3.25 to 8.92 ft. The predicted packing height varied from 3.5 ft for the 0.25 factor to 24.6 ft for the 1.75 factor.

CONCLUSIONS

Significant progress has been made, as summarized below:

- Static test results showed that the radon concentration factor for refined Mazola corn oil was in a range between 8.1 and 8.9. The

results of degassing tests conducted using heat, agitation, and ultrasonic sonication have shown that heat and agitation can remove essentially all radon from the oil, whereas the highest radon removal factor using sonication with evacuation was found to be 0.77.

- The lab-scale test apparatus having a 3-in. diameter and 16- and 42-in. high packed beds has been designed, constructed, and tested successfully. A series of dynamic radon absorption flow tests were conducted using a 1/4-in. size Raschig ring packing material in the 3-in. diameter and 42-in. packed tower. The preliminary test results showed that the radon absorption rate of 97–98.7% range was achieved for an oil rate of 259 ml/min and air rate of 0.5 ℓ/min.
- The design of a pilot engineering-scale radon removal test unit is complete. A 12-in. diameter, 80-in. high absorption column with 1-in. diameter commercial packing rings has been assembled and will be tested for Radon 220 removal.
- A computer program was modified and employed to conduct a parametric study to scope the size and operating conditions to be expected in the full commercial-scale design of a radon absorption tower for a DOE Fernald facility. The present analysis has resulted in an absorption tower with a diameter of 3.3 ft and a

tower packing height of 14 ft for 3-in. Raschig ring packing. The oil flow rate would be 268 gpm, and the gas flow rate would be 250 CFM.

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REFERENCES

- Home Buyer's and Seller's Guide to Radon*, EPA Report 402-R-93-003, March 1993.
- T. K. Sherwood, R. L. Pigford, and C. E. Wilke, *Mass Transfer*, Mc-Graw-Hill, New York, 1975.
- M. Steinberg and B. Manowitz, *Recovery of Fission Product Noble Gases*, Ind. and Eng. Chem. 51, pp. 47-50, 1959.

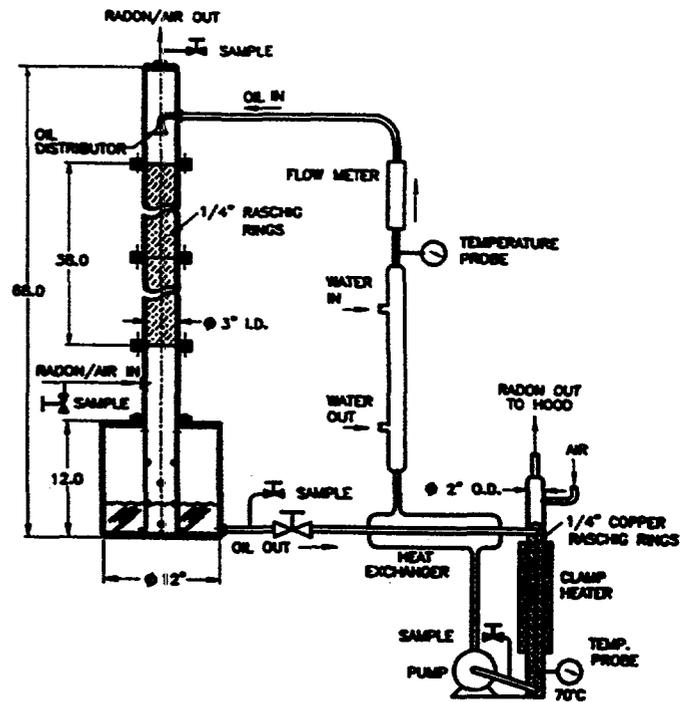


FIG. 2. THREE-IN. DIAMETER ABSORPTION / DESORPTION SYSTEM.

Concentration Factor
(Absorption Capacity / Input Concentration)

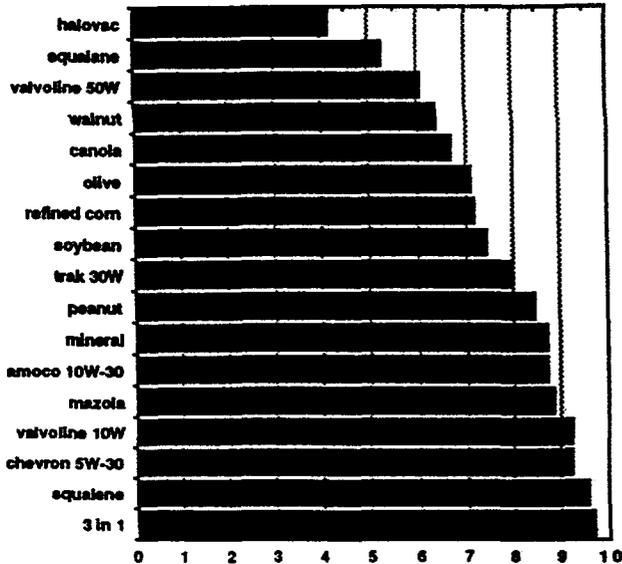


FIG. 1. HIGHEST MEASURED CONCENTRATION FACTORS OF RADON IN VARIOUS OILS AT 20°C.

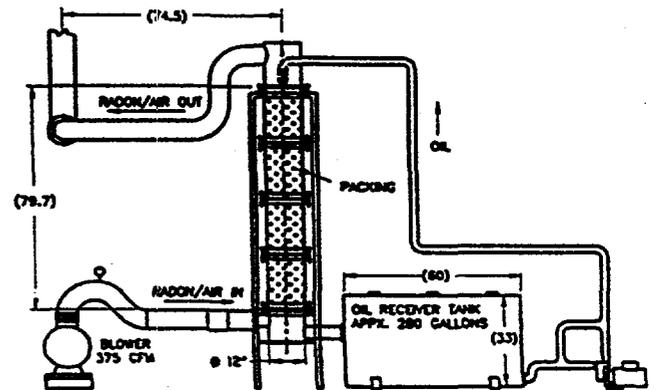


FIG. 3. TWELVE-IN. DIAMETER ENGINEERING ABSORPTION TOWER ASSEMBLY.

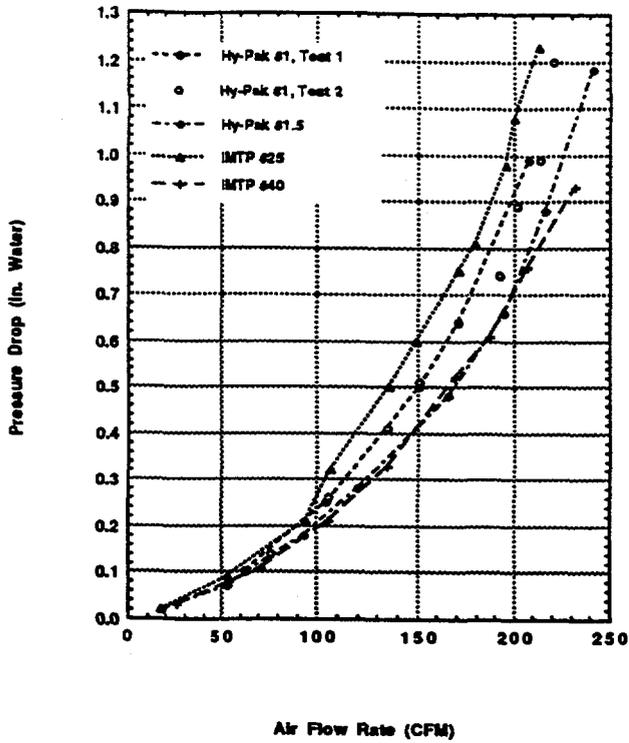


FIG. 4. PRESSURE DROP ACROSS THE 6-FT PACKED COLUMN WITH FOUR TYPES OF PACKING MATERIALS AND WITHOUT OIL FLOW

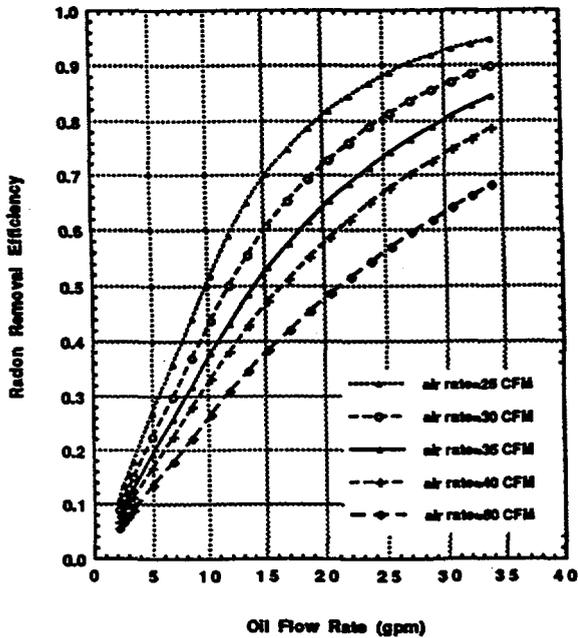


FIG. 5. EFFECT OF THE FLOW RATES ON THORON REMOVAL IN THE 12-IN. DIA. ABSORPTION TOWER WITH 1-IN. HY-PAK PACKINGS.